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■ PROGRESS REPORT NO. 9

FIRST QUARTERLY PROGRESS REPORT NO. 9

DECEMBER 16, 1964 THROUGH MARCH 15, 1965

To:

THE JOINT SERVICES TECHNICAL ADVISORY COMMITTEE

REPRESENTING: THE U.S. ARMY ELECTRONICS LABORATORIES
THE U.S. ARMY RESEARCH OFFICE
THE OFFICE OF NAVAL RESEARCH
THE AIR FORCE OFFICE OF SCIENTIFIC RESEARCH

COLUMBIA RADIATION LABORATORY NEW YORK, NEW YORK 10027

■ MARCH 15, 1965

COLUMBIA RADIATION LABORATORY ■ MARCH 15, 1965 ■ CU-00??(E)

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COLUMBIA RADIATION LABORATORY

RESEARCH INVESTIGATION DIRECTED TOWARD
EXTENDING THE USEFUL RANGE OF THE
ELECTROMAGNETIC SPECTRUM

Progress Report No. 9

First Quarterly Progress Report

December 16, 1964 through March 15, 1965

Contract DA 28-043 AMC-00099(E)
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CU-3-65 AMC-00099(E) Physics

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Physical research in fields in which microwave frequency techniques are employed; the development of microwave electronic and circuit devices.

The research reported in this document was made possible through support extended the Columbia Radiation Laboratory, Columbia University, by the Joint Services Electronics Program (U. S. Army Electronics Laboratories and U. S. Army Research Office, Office of Naval Research, and the Air Force Office of Scientific Research) under Contract DA 28-043 AMC-00099(E).

Prepared by R. Novick

COLUMBIA UNIVERSITY

Division of Government Aided Research

New York, N. Y. 10027

March 15, 1965

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P. Davidovits and W. A. Stern, "A Field-Independent Optically Pumped Rb^{87} Maser Oscillator," Appl. Phys. Letters 6, 20 (1965).

B. Budick,[4] R. Novick, and A. Lurio,[5] "Light Sources for Double-Resonance and Level-Crossing Spectroscopy," Appl. Optics 4, 229 (1965).

Papers by CRL Staff Members Presented at Scientific Meetings

American Physical Society Meetings:

Berkeley, California, December 21-23, 1964.

S. R. Hartmann, "Photon Echoes," Bull. Am. Phys. Soc. 9, 715 (1964).

New York, N. Y., January 27-30, 1965.

E. Lipworth, "Upper Bound to Electric Dipole Moment of the Electron," Bull. Am. Phys. Soc. 10, 28 (1965).

S. R. Hartmann, "Optical Excitation of Superradiant States in Ruby," Bull. Am. Phys. Soc. 10, 34 (1965).

E. B. Saloman, "Oscillator Strengths for the Lines from the $(6p7s)^3P_1{}^o$ State of Pb^{207}," Bull. Am. Phys. Soc. 10, 49 (1965).

S. Marcus, B. Budick, and R. Novick, "Level-Crossing Spectroscopy with an Electric Field; Stark Shift of the 3^2P Term in Lithium," Bull. Am. Phys. Soc. 10, 49 (1965).

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B. Budick, S. Marcus, and S. Jacobs, "Magnetic Hyperfine Interaction in an Excited State of Chromium," Bull. Am. Phys. Soc. 10, 49 (1965).

P. Davidovits and W. A. Stern, "Field-Independent Optically Pumped Rb^{87} Maser Oscillator," Bull. Am. Phys. Soc. 10, 72 (1965).

CRL Resonance Seminars

Meetings are held weekly at Columbia University, New York, N. Y. during the academic year, and are open to all members of the Physics Department. Guest speakers are invited to discuss work in the general area of the research in the Columbia Radiation Laboratory.

B. S. Mathur, Harvard University, "The Tritium Maser," January 15, 1965.

Dale Teaney, IBM Research Laboratory, Yorktown Heights, "Mn^{55} NMR and Antiferromagnetic Resonance in Cubic Antiferromagnets," February 26, 1965.

Philip C. Fisher, Lockheed Aircraft Corporation, "X-Ray Astronomy," March 12, 1965.

Lectures

I. D. Abella

"Photon Echoes," Colloquium, Brandeis University, Waltham, Massachusetts, January 6, 1965; ibid., Seminar, McMaster University, Hamilton, Canada, January 7, 1965; ibid., University of Michigan, Ann Arbor, Michigan, February 4, 1965; ibid., Colloquium, Johns Hopkins University, Baltimore, Maryland, March 4, 1965.

"Observation of Photon Echoes," Colloquium, Yale University, New Haven, Connecticut, January 13, 1965; ibid., University of Pennsylvania, Philadelphia, Pa., March 8, 1965.

"Photon Echoes in Ruby," Colloquium, Harvard University, Cambridge, Massachusetts, January 18, 1965; ibid., Seminar, University of Toronto, Toronto, Canada, January 21, 1965; ibid., University of Chicago, Chicago, Illinois, February 10, 1965; ibid., National Research Council, Ottawa, Canada, February 16, 1965.

"Photon Echoes in Ruby," Seminar, Rutgers University, New Brunswick, New Jersey, February 26, 1965; ibid., Colloquium, State University of N. Y., Stony Brook, New York, March 3, 1965; ibid., Seminar, University of Maryland, College Park, Md., March 5, 1965.

J. G. Daunt

"Isotopic Phase Separation in Solids," Colloquium, Columbia University, New York, N. Y., March 12, 1965.

P. Davidovits

"Rb^{87} Maser Oscillator," U. S. Naval Reserve Unit, New York, N. Y., December 1964.

P. Feldman

"Metastable Autoionizing Atoms," Colloquium, E. O. Hulbert Center for Space Research, U. S. Naval Research Laboratory, Washington, D. C., January 14, 1965.

S. R. Hartmann

"Photon Echoes," Seminar, University of California at Los Angeles, Los Angeles, California, December 28, 1964; ibid., University of California, San Diego, LaJolla, California, December 28, 1964; ibid., Colloquium, University of Alabama, Huntsville, Alabama, January 21, 1965; ibid., IBM Research Laboratory, Yowktown, New York, February 4, 1965; ibid., University of Wisconsin, Madison, Wisconsin, March 12, 1965.

"Nuclear Magnetic Resonance in the Demagnetized State," Colloquium, Brandeis University, Waltham, Massachusetts, February 9, 1965.

P. Kusch

"The Magnetic Moment of the Electron - A Case History in the Changing Sociology of Science," Seminar, Oregon State University, Corvallis, Oregon, January 21, 1965; ibid., University of California, Berkeley, Berkeley, Calif., February 10, 1965; ibid., University of Washington, Seattle, Washington, February 25, 1965.

"Molecular Velocity Distributions and their Application to Chemical Problem," Seminar, Stanford University, Stanford, California, February 15, 1965.

E. Lipworth

"Time Reversal Invariance and the Electric Dipole Moment of the Electron," Colloquium, Rensselaer Polytechnic Institute, Troy, New York, March 12, 1965.

S. Marcus

"Level Crossing in Lithium," Seminar, Yale University, New Haven, Connecticut, March 10, 1965.

R. Novick

"Compound Atomic States," Colloquium, National Research Council, Division of Pure Physics, Ottawa, Canada, December 18, 1964.

ABSTRACT

New measurements with greatly increased signal-to-noise ratio have been made of the hfs of the $(6p7s)^3P_1{}^o$ state of Pb-207. The level crossing was found to be at $H_C = 4661.1(4)$G. The lifetime as determined from the width of the level-crossing signal was $\tau = 5.8(2) \times 10^{-9}$ sec, while the value obtained from the Hanle effect, using a cell of separated Pb-208, was $\tau = 5.7(2) \times 10^{-9}$ sec. Saturation of the coherence-narrowing effect in lead was observed. The magnitude of the effect, assuming a Barrat-type theory, corresponds to a branching ratio to the ground state of 27% as compared with 17% obtained from Khokhlov's f-value ratios. A collision broadening cross section of $\sigma = 1.4(2) \times 10^{-12}$ cm^2 was observed in the high density range 2×10^{14} to 8×10^{14} atoms/cm^3.

Evidence has been obtained for the existence of a metastable autoionizing state in nitrogen. This state may be the $^6S^o$ state postulated as an energy storage mechanism for the pink region of the nitrogen afterglow, or it may be a new molecular metastable state.

With a He-Ne laser homodyne spectrometer, observation was made for the first time of the frequency spectrum of the time-dependent concentration fluctuation in a binary liquid mixture of cyclohexane and aniline just above the critical temperature. The frequency distribution of the scattered light was found to be Lorentzian in shape as predicted by theory. The linewidth of the scattered radiation was observed to decrease linearly with temperature at the rate of 200 ± 20 cps/$^\circ$C at a forward scattering angle $\theta = 20.5^\circ$. The linewidth also varied as $\sin^2\theta/2$.

Experimental verification has been made that pure electric field level crossings can be observed. In addition, level crossings have been observed near zero gauss by the application of a small magnetic field parallel to the electric field. These Stark effect experiments, carried out in the $^3P_1$ state of Hg, are in agreement with theory.

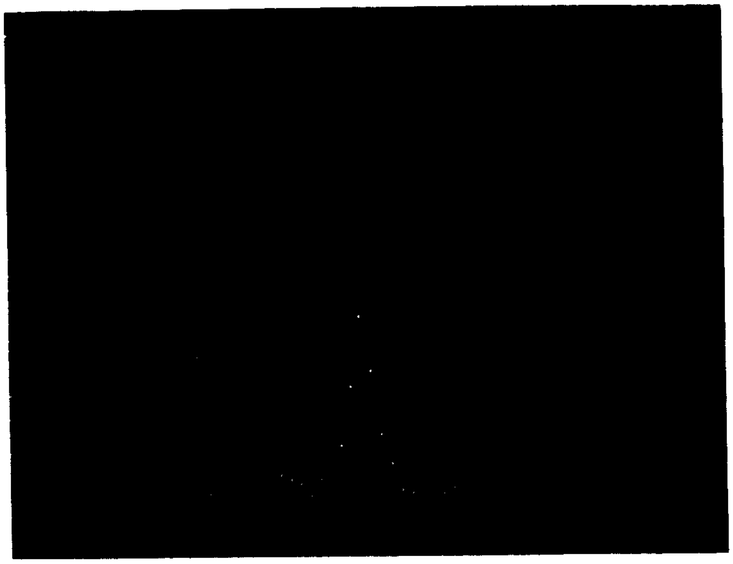

Fig. 1. Coincidence time distribution.

I. ATOMIC PHYSICS

A. PROPERTIES OF THE METASTABLE STATE OF SINGLY-IONIZED HELIUM*

1. Direct Detection of the Two-Photon Decay Mode (M. Lipeles, R. Novick, N. Tolk)

Although some improvements were made in the counting rates obtained from the fast Amperex photomultiplier tubes discussed in the last report,[(1)] real coincidence signals could not be observed despite the installation of new more stable and more sensitive electronic circuits. Therefore, the Amperex tubes were replaced by the original E.M.I. photomultipliers. Real coincidences were readily observed with these tubes. The time distribution of the coincidences was determined with a start-stop time-to-pulse height converter.

In the electronic circuits the pulses are amplified by a factor of 100 with a rise time of 2 nsec. These amplifiers limit the output at one volt with no after pulsing and virtually no ringing. In this way the large dynamic range of pulses, resulting from the low gain per stage in the electron multiplier, is compressed to the point where tunnel diode discriminators may be used. The discriminators used in our system comprise the input stages of a start-stop time-to-pulse height converter. The pulses from one channel are used to start a ramp voltage and from the other channel to stop it, thereby specifying a given output pulse height. With this system clean time-resolution curves have been obtained (Fig.1).

A set of these curves as a function of the angle between phototubes has been recorded. The plot of the data, Fig.2, shows an angular dependence having the general features of a

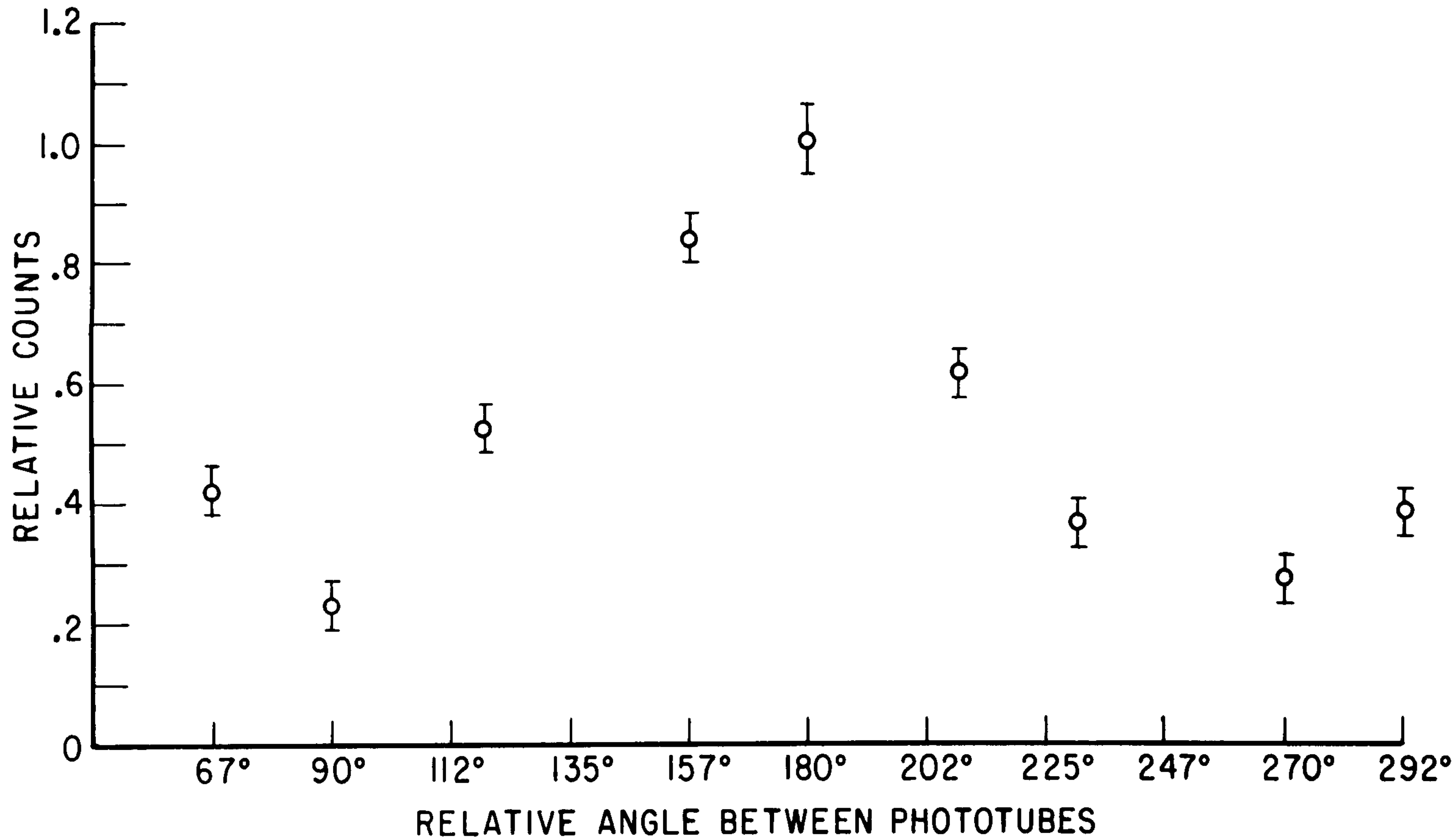

Fig. 2. Angular correlation between photons.

$(1 + \cos^2\theta)$ variation but with some distortion resulting from misalignment of the beam. Unfortunately, because of instabilities in focusing, the beam cannot be reliably positioned with respect to the detectors.

Program for the next interval: We will continue trying to improve angular dependence measurements and will attempt other tests to rule out the possibility that the observed coincidences result from spurious effects.

*This research was also supported by the National Aeronautics and Space Administration under Grant NsG-360.

(1) CRL Quarterly Progress Report, December 15, 1964, p.3.

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2. Direct Lifetime Measurements
(S. Fisher, M. Lipeles, R. Novick, N. Tolk)

Assembly of the drift tube and its accessories has begun and will continue during the next interval; new chambers will be tested.

*This research was also supported by the National Aeronautics and Space Administration under Grant NsG-360.

B. ELECTRIC-FIELD HANLE EFFECT
(W. Happer, A. Khadjavi, A. Lurio)

During this quarter further studies were made of the feasibility of using zero-field level-crossing techniques to measure the Stark shift of excited atomic states. Evaluations have been made of the relative merits of level crossing in a pure electric field and of level crossing in parallel electric and magnetic fields.

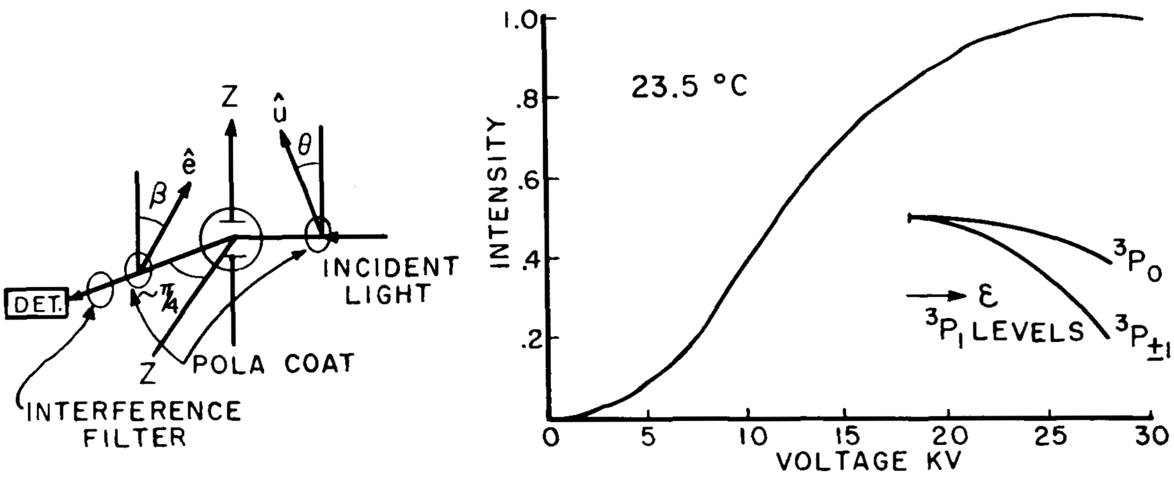

Fig. 3. Pure electric-field Hanle effect.

Figure 3 shows the experimental details of the pure electric-field Hanle effect. Optimum signal is obtained when both the incident and the observed radiation are polarized at 45° to the electric field. The signal also depends on the angle between the incident and detected beams of light, reaching a maximum for forward or back scattering and going to zero at 90°. The form of the signal,

$$I \propto \frac{1}{1 + [(\alpha E^2 \tau)/\hbar]^2} , \tag{1}$$

is independent of the angle between the beams — in contrast to magnetic-field Hanle effect. It is important to note that in the pure electric-field level crossing one must know the effective lifetime of the excited states in order to extract α. As is well known, this can be strongly influenced by coherence narrowing and collisions. By use of the accepted lifetime for the $^3P_1$ state of mercury[1] and our pure electric-field Hanle effect data, the value of α obtained is about half that measured by Blamont. This may be due to uncertainty in the effective lifetime or to the presence of uncancelled magnetic fields.

In order to avoid complications due to changes in the effective lifetime of the excited states, parallel electric and magnetic fields have been used. The geometry is shown in Fig. 4. A cell containing the sample to be studied was illuminated with resonance radiation along the y axis and subjected to axial electric and magnetic fields. Analysis of the data showed the presence of some odd-isotope crossings as well as the even-isotope crossings. The intensity of scattered resonance radiation along the x axis is given by:[2]

$$I_{even} \propto \sin^2\theta \, \sin^2\beta \cdot \frac{1}{1 + [(2\delta H)/\Gamma]^2} - \tfrac{1}{2} \sin 2\theta \, \sin 2\beta$$

$$\times \left\{ \frac{(\delta H + \Delta\omega)/\Gamma}{1 + [(\delta H + \Delta\omega)/\Gamma]^2} + \frac{(\delta H - \Delta\omega)/\Gamma}{1 + [(\delta H - \Delta\omega)/\Gamma]^2} \right\}$$

$$I_{odd} \propto \sin^2\theta \, \sin^2\beta \left\{ \frac{1}{1 + [(2\delta H + \Delta\omega)/\Gamma]^2} + \frac{1}{1 + [(2\delta H - \Delta\omega)/2]^2} \right\}$$

$$- \sin 2\theta \, \sin 2\beta \left\{ \frac{(\delta H + \Delta\omega)/\Gamma}{1 + [(\delta H + \Delta\omega)/\Gamma]^2} \right.$$

$$\left. + \frac{(\delta H - \Delta\omega)/\Gamma}{1 + [(\delta H - \Delta\omega)/\Gamma]^2} \right\} .$$

Here θ and β are the polarizations of the incident and scattered light respectively, and $\delta H = (1/\hbar) g_J \mu_o H$ and $\Delta\omega = (1/\hbar)\alpha E^2$ are the Zeeman and Stark energies. Our results indicate a value for the Stark shift of the $^3P_1$ state of mercury of

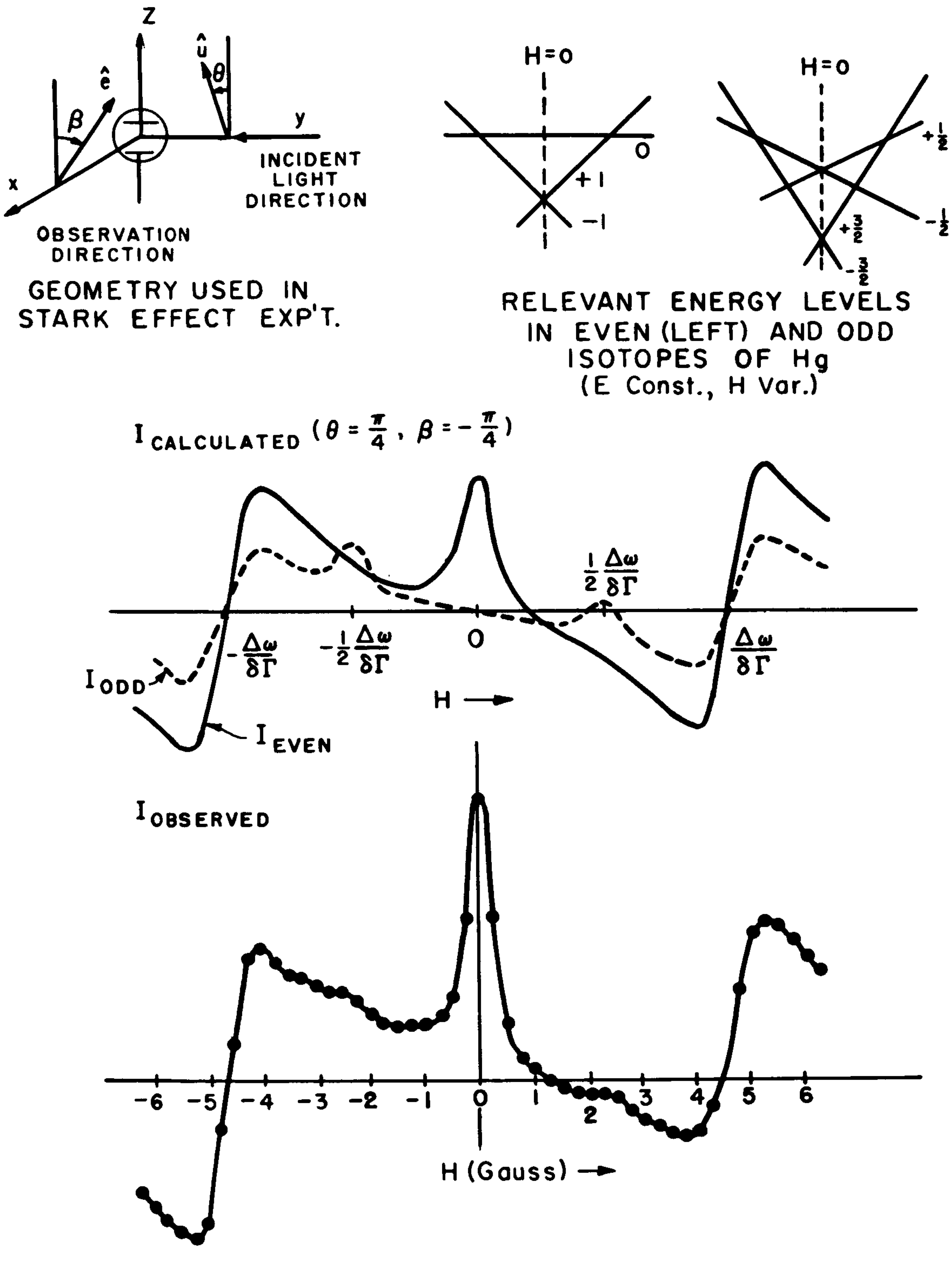

Fig. 4. Zero-field level crossing, parallel electric and magnetic fields.

$$\alpha/h = 1790 \pm 270\ cm^2/sec(kV)^2$$

At present we are improving our experimental arrangement to obtain more accurate values for the mercury Stark shift. Further, we are trying various designs of cells for Cd and Zn. We plan to extend this experiment to alkalis in the future.

(1) J. E. Blamont, Ann. Phys. (Paris) 2, 551 (1957).
(2) M. E. Rose and R. L. Carovillano, Phys. Rev. 122, 1185 (1961).

C. FINE STRUCTURE OF SINGLY-IONIZED LITHIUM*
(W. Kahan, T. Lucatorto, R. Novick)

Preliminary to measuring the fine structure of the $2\,^3P$ state of singly-ionized lithium, our efforts have been devoted to measuring the Zeeman hfs of this state by a resonance method. The predicted signal-to-noise ratio for such a resonance is the order of unity. The interaction between the electric-field component of the radio-frequency radiation and the plasma has prevented us so far from observing any Zeeman lines. During this quarter we installed and tested a new rf loop which encloses the interaction region in an equipotential box of silver. This design is intended to reduce the rf electric field without any reduction in the rf magnetic field. Preliminary tests with hydrogen indicate that this design successfully eliminates objectionable plasma effects. It has been necessary to build a molybdenum replacement to this loop, because at an operating temperature of 400°C, which is necessary to prevent lithium condensation, the lithium amalgamated with the silver and produced a structural failure. Although the resulting damage to the system has temporarily curtailed our work with lithium, an intensive program will resume in about two weeks.

Program for the next interval: By use of the new type of rf source, observation of the rf induced Zeeman hfs transitions is anticipated. If this source produces an electric-field component which, although much smaller than the previous source, still produces undesired plasma resonances, we shall investigate additional methods for eliminating this effect.

If with the above improvements the Zeeman hfs lines are not found, we shall turn our attention to development of the necessary microwave equipment (60-90 kMc) to produce radiation suitable for inducing fine structure transitions. Here the probability for success is much higher since a resonance will be accompanied by a greater change in polarization.

*This research was also supported by the National Aeronautics and Space Administration under Grant NsG-360.

D. METASTABLE AUTOIONIZING ATOMS
(L. M. Blau, P. Feldman, M. Levitt, R. Novick, G. Sprott)

1. Metastable Alkali Atoms

In the previous Quarterly Report[(1)] the results were given of a Stern-Gerlach type of experiment on metastable potassium atoms. Since then, the deflecting magnet has been calibrated and the same experiment run on metastable cesium atoms.

Calibration was accomplished by measuring the deflection of ground-state $^2S_{\frac{1}{2}}$ potassium atoms with a movable hot-wire detector. At moderately high magnetic fields, the two magnetic components ($M_J = \pm\frac{1}{2}$) in the beam appeared as completely separated peaks symmetrically spaced about the center of the undeflected beam. The data were fitted to the calculated deflected beam profiles in order to determine the value of the magnetic field gradient as a function of magnet current. The maximum gradient obtained was 1.0×10^5 G/cm.

Use of this calibration to interpret the data on potassium showed that the observed Stern-Gerlach pattern could not be fit by assuming a single magnetic sublevel to be the dominant component of the beam. The average value of a magnetic moment which would give the same beam width at half-maximum intensity as the observed data is about 3 Bohr magnetons. Moreover, this average value appears to be larger at the highest field gradients possible. This latter effect may be instrumental since at very high fields the Zeeman quenching is of the order of 80% and the signal-to-noise ratio is very poor. In any case, the data remain consistent with the assignment of the state as $(3p^5 4s3d)\,^4F_{9/2}$ and with the characteristic of differential metastability.

The Stern-Gerlach pattern of metastable cesium atoms was also measured in an attempt to determine the nature of the two observed thresholds. As noted before,(1) one of these peaks corresponds to a state with a lifetime ≈ 40 μsec, while the other has an apparent lifetime greater than 1 msec (the upper limit of sensitivity to change in signal as a function of distance from the source). Other experiments indicate that this latter signal arises from photons, and in the present experiment no deflection was observed at the highest field gradients. Alternatively, the other state did produce an observed deflection, though not as large as for the corresponding gradients in the case of potassium. The interpretations of the data from cesium still remain an open question.

At present, an experiment to measure hfs in the longer-lived alkali metastable states by means of a Rabi-type radio-frequency technique is being designed. This experiment should yield positive identification of all of the observed metastable states to date and perhaps provide a definitive explanation of the cesium data.

2. Rf Spectroscopy

In the last Quarterly Progress Report[2] it was stated that the previously unsuccessful search for Zeeman transitions in the $(1s2s2p)\,^4P_{5/2}$ state of Li^7 would be extended to fields of several kilogauss, before attempting to observe direct hyperfine transitions. A subsequent extension of our theoretical results has indicated, however, that the latter experiment is more feasible. This is due to the following considerations.

A simplified theory was postulated for an experiment[3] in which the metastable beam was quenched by passing it through moderately strong magnetic fields (up to 18 kG). This gave an approximate expression for the lifetime of each metastable state in terms of the amplitudes of its J=5/2, 3/2 and 1/2 components. This result, coupled with the information that even at 3 kG, the relative J=3/2 admixtures of adjacent Zeeman levels differ by only one part in 10^5, indicated that these Zeeman levels of the $^4P_{5/2}$ state have undetectably different lifetimes. Consideration of Zeeman transitions at much higher magnetic fields requires a difficult alteration of the Bendix multiplier or the installation of a new detector.

On the other hand, each $^4P_{5/2}$ hyperfine state with quasi-good quantum number F=3 was found to have a 2% admixture of J=3/2, which was relatively independent of magnetic-field value or trial wave function. These states were predicted to have appreciably different lifetimes (about 10%) from F=4 states, which are pure J=5/2. The uncertainty in the hyperfine splitting for states of the same M_F, corresponding to a reasonable range of Li wave functions, is about 0.3 Gc. For each wave function used, however, the difference in the splitting between two different pairs of hyperfine states having $M_F=M$ and $M_F=M \pm 1$, respectively, is only 100 Mc. To reduce the time needed to search through such a 100-Mc-wide region, it is planned to broaden the output spectrum of an Eimac X-1149 reflex klystron to about a 10-Mc width by using a General Radio 1390-B random noise generator. The necessary microwave circuitry is presently being built.

Calculations also indicate that the decrease in the total metastable signal at resonance will be comparable to the experimentally observed noise level. To observe such resonances, the time constant of the presently used lock-in amplifier has been increased to 100 seconds. Work has also begun on obtaining suitable pulses from the Bendix multiplier in order to count the individual metastables. This work will be actively pursued during the next quarter.

3. The Sextet Nitrogen Atom

The possibility of metastable autoionizing atomic states in atoms other than the alkalis was considered previously.[(4)] The general requirement for such a state is that it should lie above the ionization potential and have a multiplicity (total spin quantum number) greater than that of any of the normal atomic states of the atom. Moreover, excitation should not require a double electron impact from the ground state as this would greatly reduce the production cross section.

A particular case of interest in connection with the afterglow produced by an electrical discharge in nitrogen is the $^6S^o$ state of the $(2s2p^33p)$ configuration. This state has been postulated(5) as a possible energy storage mechanism and has been predicted to lie at 17.2 eV, 1.6 eV above the ionization potential of nitrogen. To date there has been no experimental evidence(6) to support this hypothesis, even though a second "critical potential" for ionization of nitrogen molecules was observed to result from collisions of N_2^+ ions on neutral N_2.(7)

The apparatus was modified to allow molecular nitrogen to enter the system through a Granville-Phillips variable leak. A copper tube connects the gas inlet with the well of the oven used in the alkali experiments so that a beam of N_2 has the same geometry as previously. An excitation function, shown in Fig. 5, was obtained using a 100-kc heated tungsten filament and the modified Bendix electron multiplier detector. Thresholds at $\approx$10 eV and $\approx$30 eV were observed. Allowing 9.8 eV for dissociation energy, the latter could correspond to the metastable sextet state.

The signal above 30 eV was found to be sharply reduced by a moderate electric field placed just in front of the detector transverse to the beam path. A curve of observed signal vs voltage applied across the two deflecting plates is shown in Fig. 6.

While the electric-field quenching was quite unexpected (a small amount of quenching was also observed with a strong external magnetic field), it is not at all inconsistent with the

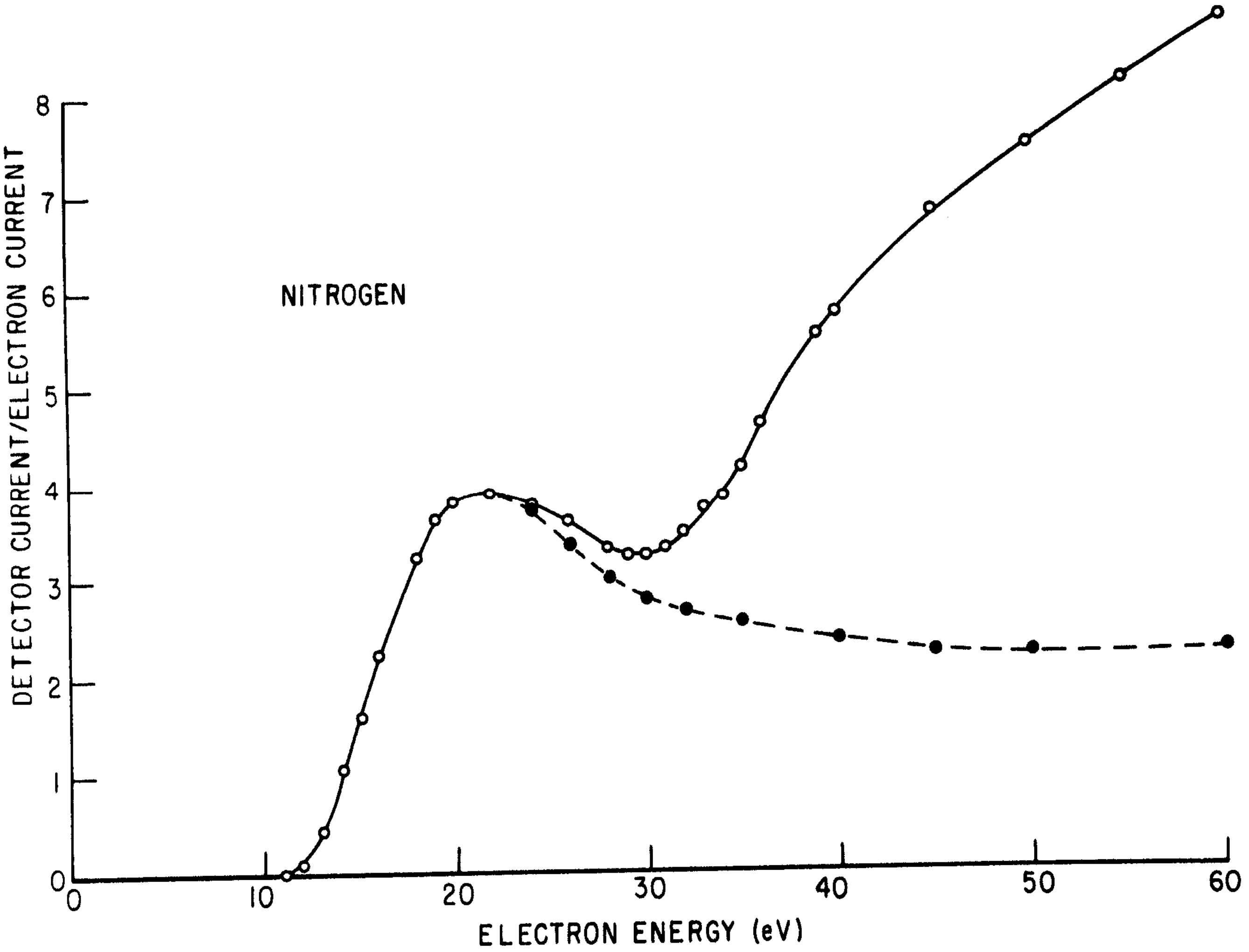

Fig. 5. Excitation curve for molecular nitrogen excited by electron bombardment. This curve was obtained using the Bendix multiplier detector. The dotted curve is the excitation function for the known metastable molecular states.

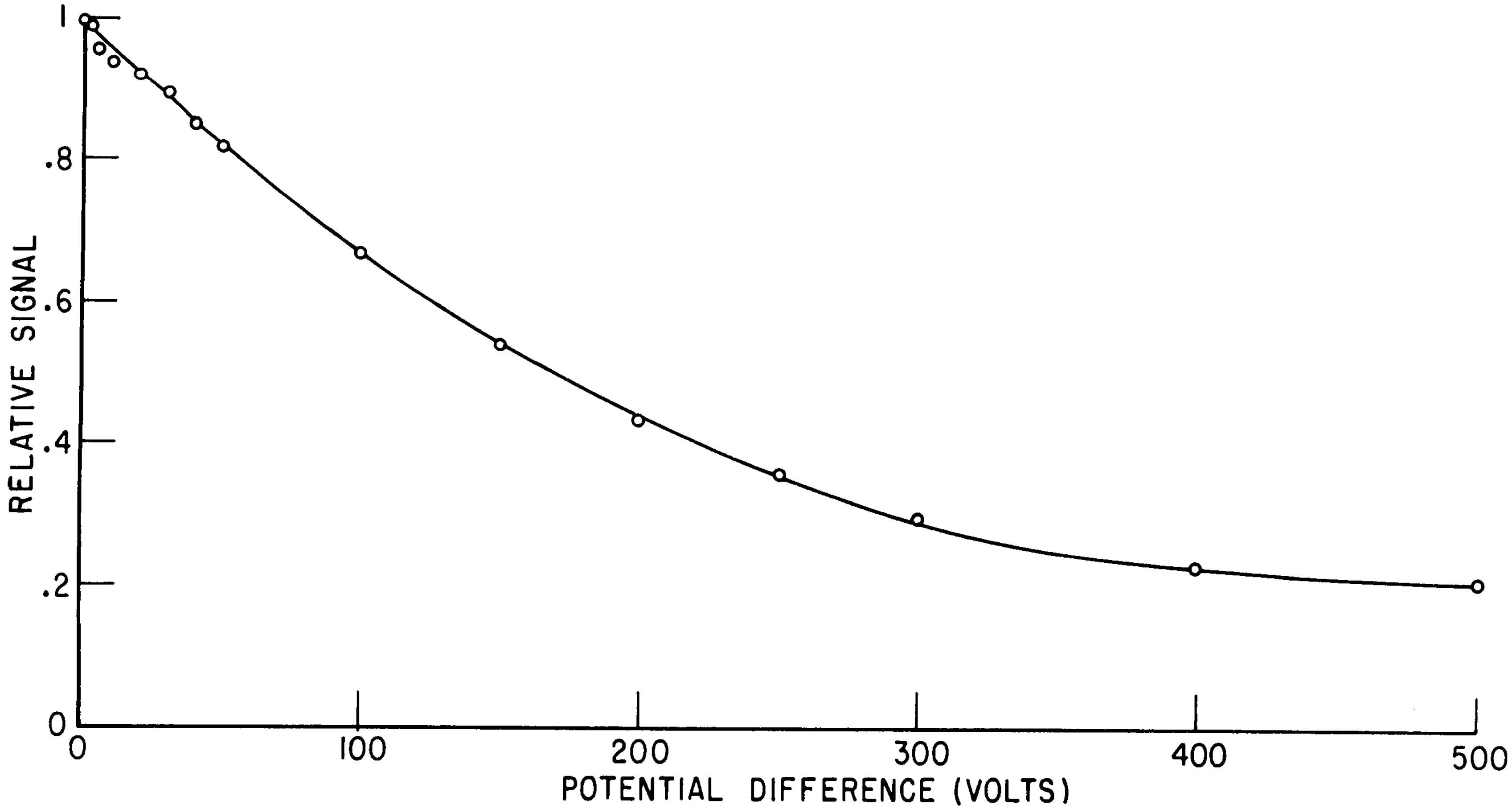

Fig. 6. Metastable signal from nitrogen as a function of voltage applied across quenching plates. The bombarding energy was 60V.

properties of the sextet state. In fact, on the basis of afterglow studies, the expected lifetime of the state is of the order of milliseconds so that in the present apparatus very few metastable atoms would autoionize and be detected. If, however, the state is quenched at moderate electric fields, then the detector itself will quench atoms in this state and the decay products will be readily observed. An attempt to use the movable detector consisting of a single collecting plate biased at -50 V was unsuccessful, probably because of the small magnitude of the electric field.

The low energy threshold has been identified as the metastable $a^1\Pi_g$ and $A^3\Sigma_u^+$ molecular states.[8] The signal arises from Auger electrons produced by collision of a small fraction of the metastable molecules with the surfaces in the detector. The identification was made from data taken with an oxide-coated cathode instead of the tungsten filament in order to obtain an energy resolution of less than 0.4 eV, and comparison was made with the published excitation curve of Lichten.[8]

<u>Program for the next interval</u>: The data taken to date do not provide conclusive evidence for the sextet nitrogen atom, and in fact the signal may indicate the existence of an autoionizing molecular state. A mass spectrometer which will analyze only ions resulting from autoionization is being installed in an attempt to solve this problem. Also, the use of an oxide-coated cathode now permits measurements of the threshold energies of the previously observed metastable state in the alkalis to within 0.2 eV, and the absolute energy can be determined by calibration with the known energies of the molecular metastable states of nitrogen.

*This research was also supported by the National Aeronautics and Space Administration under Grant NsG-360.

(1) CRL Quarterly Progress Report, December 15, 1964, p. 7.

(2) CRL Quarterly Progress Report, December 15, 1964, p. 13.

(3) CRL Quarterly Progress Report, December 15, 1963, p. 17.

(4) P. Feldman, Thesis, Columbia University (1964) p. 120 (unpublished).

(5) A. B. Prag and K. C. Clark, J. Chem. Phys. 39, 799 (1963).

(6) R. A. Young, R. L. Sharpless, and R. Stringham, J. Chem. Phys. 40, 251 (1964).

(7) N. G. Utterback and G. H. Miller, Phys. Rev. 124, 1477 (1961).

(8) W. Lichten, J. Chem. Phys. 26, 306 (1957).

E. HYPERFINE STRUCTURE OF GROUP IIA ISOTOPES*
(R. J. Goshen, R. Novick, M. Swagel)

Another natural barium cell was tested during this quarter. The maximum $6^1P_1 - 6^1S_0$ transition Hanle effect signal was obtained with S/N = 1000 at a temperature of 600°C, while the maximum $6^3P_1 - 6^1S_0$ transition Hanle effect signal was observed with S/N = 2 at a temperature of 750°C. In our previous run in June 1964, the maximum singlet transition Hanle effect signal was obtained at 400°C and the triplet Hanle effect signal at 600°C.

The magnesium oxide windows became clouded with barium after several hours of operation at a cell temperature of about 600°C. The windows are heated only on their edges, where they touch the hot metal cell. Magnesium oxide is a poor conductor of heat, and the windows are consequently much colder than the metal cell body. It was suspected that the obscuration could be reduced by an improved method of heating the windows. Therefore, a lattice structure has been added to each of the window flanges to provide radiative heating over the entire face of the crystal windows (Fig. 7).

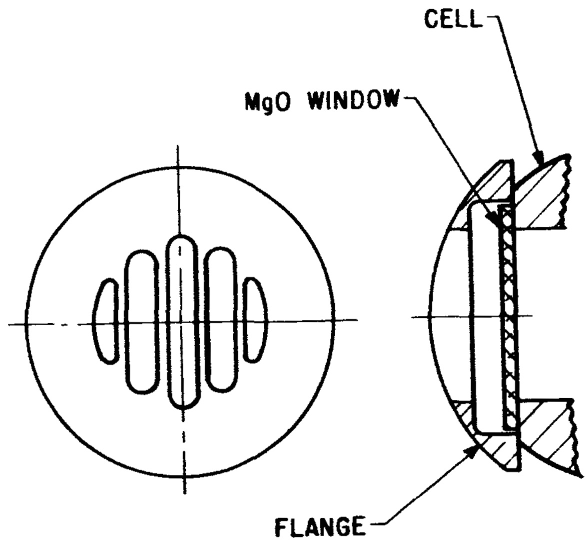

Fig. 7. Latticed window flange.

Another barium cell is now being prepared and will be tested shortly. It was thought that the molybdenum cell was reacting with barium at 600°C; therefore the use of molybdenum was discontinued. An improved stainless-steel 305 cell is being constructed. This cell should have a better vacuum seal than the previous cells. Also the 305 stainless steel retains a magnetic permeability of less than 1.01 even after considerable machining.

Program for the next interval: The natural barium cell now being prepared will be tested. At present, lamp fluctuations are limiting the S/N ratio in the experiment. A titanium trap has been installed in the argon gas-handling part of the hollow cathode lamp to improve stability and is now being tested. The next barium cell will have the latticed window flanges.

*This research was also supported by the National Aeronautics and Space Administration under Grant NsG-360.

F. FINE AND HYPERFINE STRUCTURE OF THE 3P STATE OF Li^7*
(R. C. Isler, S. Marcus, R. Novick)

During this quarter, the work toward a higher precision measurement of the fine structure of the 3P state of Li^7 was continued. Our major effort was directed toward improving the homogeneity of the magnetic field. By means of painstakingly adjusting the spacing of the pole pieces, a homogeneity better than 1 part in 10^5 over a region 2 in. $\times$ 2 in. $\times$ 1 in. (1 in. in the direction parallel to the magnetic field) was obtained.

In an attempt to improve the stability of the magnetic field, a high-stability Magnion power supply HS-1365B was purchased. Preliminary tests indicate a short-term magnetic-field stability of a few parts per million, a significant improvement.

A study was made to determine the optimum value of modulation current. An rms current of 0.4 A was found to give maximum signal resolution with no serious loss in signal-to-noise ratio.

Program for the next interval: We plan to continue our efforts toward a precision measurement of the fine structure of the 3P state of Li^7 and to continue the study of the Stark effect.

*This work was also supported by the National Aeronautics and Space Administration under Grant NsG-360.

G. ELECTRON SCATTERING SPECTROMETER
(P. Feldman A. T. Kung, R. Novick)

In a previous Quarterly Progress Report[1] a new experimental program for the study of resonances in the elastic scattering cross sections of atoms was described. The original experimental arrangement was designed about an electrostatic electron trap which would permit analysis of electrons scattered from a beam of atomic hydrogen.

Toward the end of 1964, the motivation for performing the experiment with atomic hydrogen was removed. Schulz[2] reported the observation of a weak resonance in H at about 9.6 eV, and the discrepancy[1] in theoretically predicted values for this resonance was also resolved. Hence the need for an electrostatic electron trap, the most difficult experimental problem in this project, was obviated. The present experimental design achieves a large scattering volume and long electron time-of-flight by means of a long path length ($\approx$1 to 2 m) from electron source to detector. A vapor of the material under investigation will fill the entire volume of the apparatus. Energy analysis of the electron beam will be accomplished by a measurement of the spread in arrival time at the detector of an initially narrow ($\approx$1 nsec) electron pulse.

The advantage of a time-of-flight spectrometer over electrostatic analyzer spectrometers[3] is its insensitivity to elements such as the alkalis or the alkaline earths which affect the surface potentials and hence the energy resolution of electrostatic devices. The technical difficulty in constructing a time-of-flight apparatus arises in the formation and detection of very short electron pulses, but this should lie within the present state of the art of pulsed electronics techniques.

A large number of experiments may be performed with the apparatus now being designed. These include: 1) resonances in the elastic scattering cross section due to negative ion formation in the alkalis, alkaline earths, and Group II elements such as zinc, cadmium, and mercury; 2) inelastic excitation of very short-lived autoionizing states in these same elements; 3) inelastic excitation of various vibrational levels in excited electronic states of molecules from which potential energy curves of these states may be deduced; 4) inelastic excitation of autoionizing electronic states in molecules.

Program for the next interval: The vacuum system previously described(1) is being assembled to allow for testing of various pulsed electron sources. A long stainless-steel drift tube and the electronic circuitry are being designed. Calculations are in progress to determine the maximum energy resolution possible under normal experimental conditions.

(1)CRL Quarterly Progress Report, September 15, 1964, p. 23.

(2)G. J. Schulz, Bull. Am. Phys. Soc. 10, 184 (1965).

(3)G. J. Schulz, Phys. Rev. 125, 229 (1962).

H. ELECTRIC DIPOLE MOMENT OF THE CESIUM ATOM
(E. Lipworth, P. G. H. Sandars)

The apparatus referred to in the previous progress report is now complete. Resonances have been observed in cesium in glass and quartz bulbs with various wall coatings and also in uncoated glass bulbs filled with a neon buffer gas.

Serious difficulties have been encountered. When a

20-cps electric field of magnitude $\approx$2000 V/cm is applied across the gas-filled cells the resonance immediately disappears, presumably because a surface film of cesium is burnt off the glass walls by oscillating surface currents. The walls then act as a pump for cesium atoms, lowering the effective cesium density in the bulb. The resonance can be brought back by heating the bulb and maintained even in the presence of the electric field, but gas breakdown prevents the application of a field large enough to observe even the quadratic Stark shift between the cesium Zeeman levels.

Even more unpleasant effects occur in vacuum bulbs with coated walls. With a Paraflint wax coating a 20-cps (1000 V/cm) electric field produces a large intensity modulation of the transmitted light at a frequency of 40 cps. This 40-cps signal is larger than the resonance signal at 20 cps and is probably produced by wall heating effects which change the cesium density at twice the modulation frequency (heating is proportional to E^2). A 40-cps signal would not in itself be objectionable because one could discriminate against it by lock-in techniques; however, it is noisy and accompanied by a small signal at 20 cps whose origin is not understood. The situation is improved when bulbs coated with Dri-Film are used. The second harmonic signal is still present but smaller and much less noisy.

In view of these difficulties and the fact that the principal investigator is only temporarily at Columbia Radiation Laboratory, work on this project is being suspended. It may be continued at a later date.

A. OPTICAL STUDIES OF RADIOACTIVE ATOMS

(W. Happer, R. Novick, E. B. Saloman)

The level-crossing effect was remeasured in the $(6p7s)\,{}^3P_1^o$ state of Pb^{207} after the experimental setup had been optimized for maximal signal-to-noise ratio. The λ = 2833-Å line was used to detect the effect. A typical piece of data is shown in Fig. 8. From observation of a total of 43 resonances, the level crossing was determined to be at H_c = 4661.1(4) G. The quoted uncertainty is twice the statistical uncertainty. The level-crossing signal was observed for all three fluorescent lines. The intensity and signal-to-noise ratio were best for the 3639-Å line, almost as good for the 2833-Å line, and considerably poorer for the 4058-Å line. The 4058-Å fluorescence

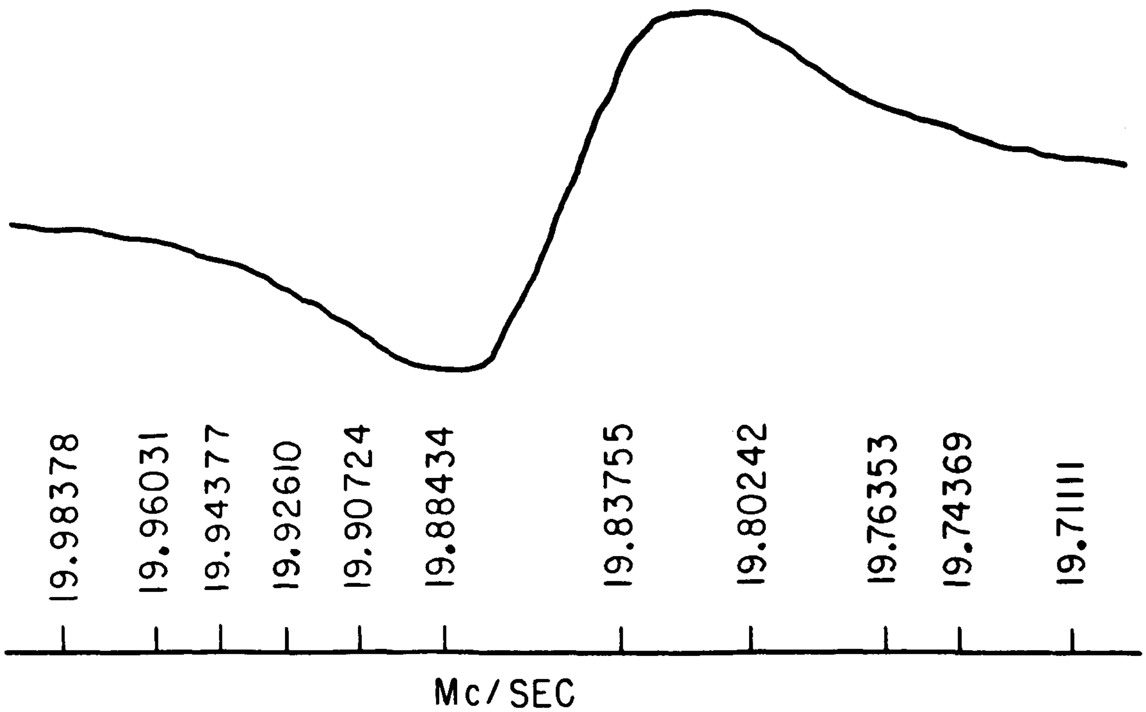

Fig. 8. High-field level-crossing data.

(terminal state J=2) should be less polarized than the 3639-Å and 2833-Å lines (terminal states J=1 and J=0 respectively), but the large branching ratio to the J=2 state implied by Khokhlov's f-value ratios[1] should compensate for the smaller degree of polarization. Although the detection optics may account for some of the difference, it seems that the accepted branching ratio to the J=2 state is too large.

The level-crossing effect was next used to determine the lifetime of the $(6p7s)\,^3P_1^o$ state. Since only one isotope of natural lead has a high-field level crossing, there can be no confusion from mixtures of the signals from the even isotopes with that from the odd isotope as was possible in our previous determination. Because of the large isotope shifts of the various lead isotopes, coherence-narrowing effects can occur only between atoms of the same isotope. Since only 21% of the atoms in the cell were Pb^{207}, we would roughly expect $(0.21)^2=1/25$ of the magnitude of the coherence-narrowing effect as observed in the 100% Pb^{208} cell discussed below. Thus, less than a one-percent effect would be expected. This was confirmed experimentally by observing the width of the level-crossing signal at temperatures corresponding to a change in density of about ten. No change was observed to within the statistical uncertainty of about 2%. The observed widths were plotted against the square of the modulation as shown in Fig. 9. When extrapolated to zero modulation and corrected for the off-centered NMR probe, the width of the level-crossing signal is observed to be 12.6(4) G, which corresponds to a lifetime of $5.8(2) \times 10^{-9}$ sec.

The lifetime was then redetermined by the Hanle effect using a cell which contained 99.75% Pb^{208} and thus eliminated any confusion with the odd-isotope Hanle-effect signal.

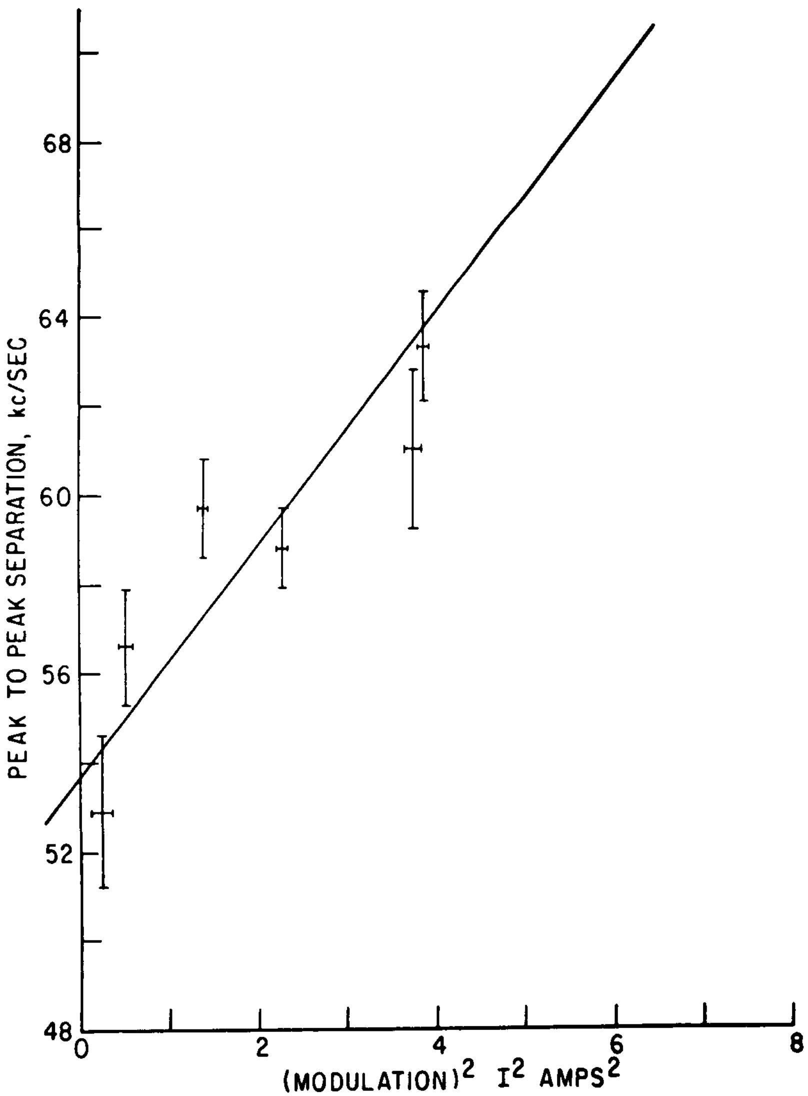

Fig. 9. High-field level-crossing data peak-to-peak separation vs modulation squared.

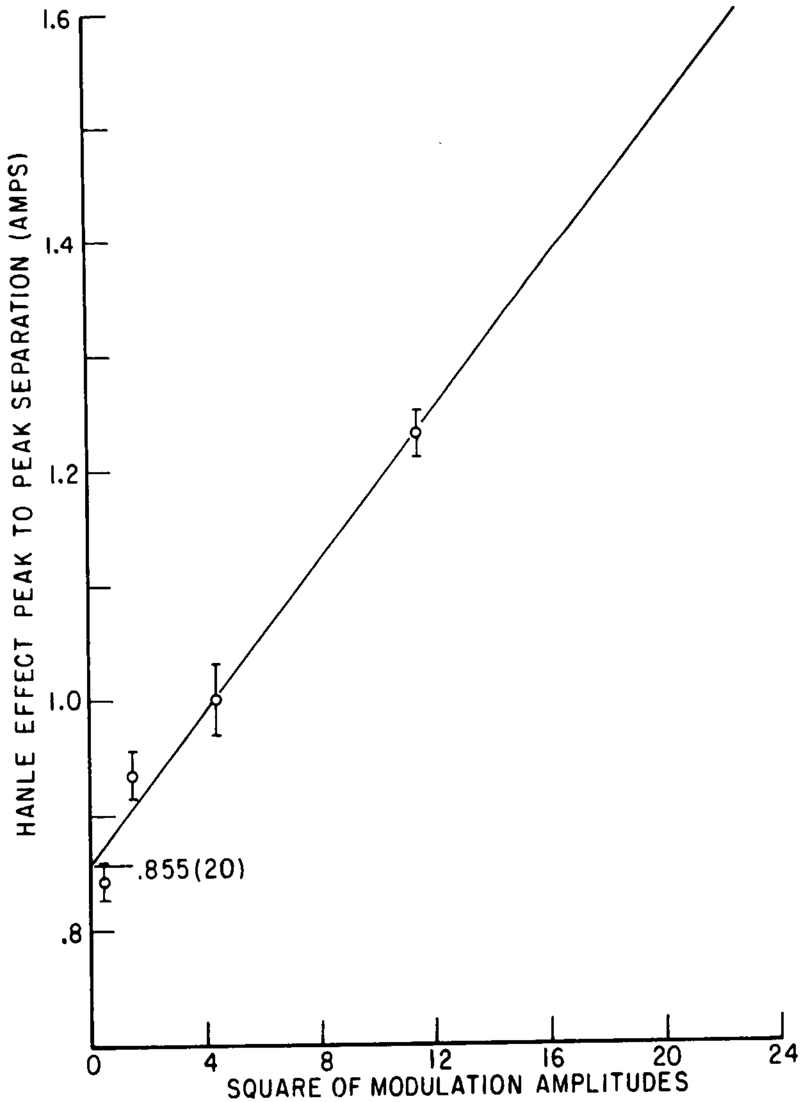

Fig. 10. Hanle effect data: peak-to-peak separation vs modulation squared.

However, the concentration of one single isotope made the coherence-narrowing effect particularly pronounced. This effect was compensated for by lowering the atomic densities to 1.8×10^9 atoms/cm^3 in the 50-cm^3 cell. A plot of peak-to-peak separation versus modulation is shown in Fig. 10, while the peak-to-peak separation-vs-density curve is shown in Fig. 11. From this and our Helmholtz coil calibration of 10.122 G/A, we obtain a peak-to-peak separation at low density of 8.6(3) G, which corresponds to a lifetime of $5.7(2) \times 10^{-9}$ sec. Both this value and the one obtained from the high-field level crossing are in good agreement with our previous determination $\tau = 5.8(5) \times 10^{-9}$ sec, which was obtained by Hanle-effect studies on natural lead with a correction estimated for the presence of the odd isotope. Use of the new silver cell oven allowed the cell to be uniformly heated.

To our knowledge, Fig. 11 demonstrates the first observations of the saturation of coherence narrowing, which was predicted by Barrat.[2] These observations were made possible by the presence of branching in lead. In an atom with only one decay branch, the large number of scatterings necessary to insure saturation results in almost complete depolarization and spatial isotropy of the escaping light. Since level-crossing and optical double-resonance techniques monitor changes in polarization and angular distribution of the scattered light, these measurements become extremely difficult when strong trapping of resonance radiation occurs and no branch mode is available to allow the photons to escape. Although the signal from the 2833-Å line does approach zero at high densities, the signals from the 3639-Å and 4058-Å lines are essentially unaffected. To check that no appreciable buildup of metastable atoms

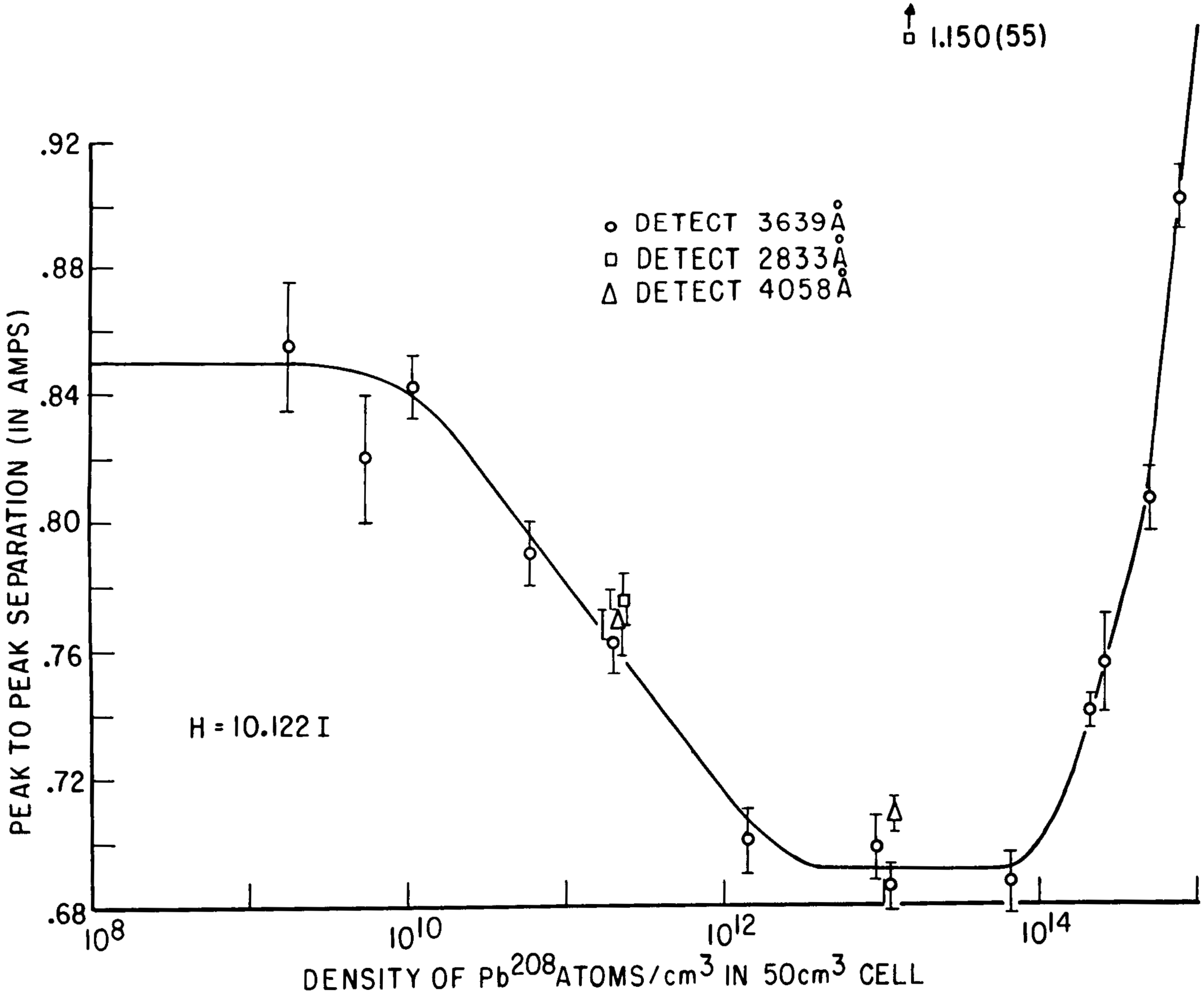

Fig. 11. Experimental Results: peak-to-peak separation vs density.

resulted from the branch fluorescence, the width of the Hanle-effect pattern was measured for different incident light intensities. No change was observed in the linewidth for input intensities differing by a factor of ten.

The theory of coherence narrowing[2] may be extended to the case of excited atomic states with several fluorescent decay modes in addition to the resonant line to the ground state. The density matrix for an ensemble of atoms excited at t=0 is then

$$\rho(t) = e^{-\Gamma\left(1-\frac{\Sigma x_i \alpha_i \Gamma_i}{\Gamma}\right)t}\left[e^{-i\mathcal{H}t}\rho(0)\, e^{i\mathcal{H}t} - \frac{II}{2F+1}\right] + e^{-\Gamma\left(1-\frac{\Sigma x_i \Gamma_i}{\Gamma}\right)t}\frac{II}{2F+1} \tag{1}$$

The various decay branches are labeled by their branching ratio Γ_i/Γ, the reabsorption probability for the branch photon x_i, and the depolarization factor α_i. The notation is identical to that employed by Barrat,[2] and Eq. (1) reduces to his standard form for the case of only one branch. The detected intensity in the i^{th} branch is then

$$I_i(\lambda)\Delta\Omega = \Delta\Omega Z h\nu_i(1 - x_i)\ 2\pi|Ak_o|^2\ \rho(k_o) \sum_{\mu_i m' m''} \langle\mu_i|\hat{\ell}_\lambda \cdot \vec{D}|m'\rangle$$

$$\times\ \left[\int_0^\infty dt\ e^{-im'\omega t}\ \langle m'|\rho(t)|m''\rangle\ e^{im''\omega t}\right]\ \langle m''|\hat{\ell}_\lambda \cdot \vec{D}|\mu_i\rangle, \tag{2}$$

where Z is the excitation rate. From Eqs. (1) and (2) it is evident that the measured lifetime for all branches is given by

$$T = \frac{\tau}{1 - (\Sigma x_i \alpha_i \Gamma_i)/\Gamma} \tag{3}$$

where $\tau = 1/\Gamma$ is the real radiative lifetime.

When no branching is present, complete depolarization of the scattered light occurs as the reabsorption probability approaches unity. This accounts for the difficulty of observing the full predicted coherence narrowing in atoms with a single decay mode. When branching is present the fluorescent lines can exhibit a sizable polarized component even when the reabsorption probability for the resonant line approaches one. The transparency of the vapor to fluorescent photons thus makes atoms with branch decay modes ideally suited for studies of depolarization effects at very high vapor densities. If the above theory is correct, the observed coherence narrowing corresponds to a branching ratio of 27% to the ground state as compared with 17% predicted by Khokhlov's f-value ratios. The predictions of this theory for a 27%-branching ratio with Barrat's parameter L=1 and with our lifetime value are shown in Fig. 12 at the densities less than 10^{13} and may be compared with the experimental results in Fig.11.

The widths of the various decay lines from the $^3P_1^o$ state were compared at two values of atomic density, one on the slope of the coherence-narrowing curve and one where the coherence-narrowing curve saturates. At the lower density, (2×10^{11} atoms/cm^3) on the slope of the curve, all three decay modes gave the same width Hanle-effect signal to within statistical uncertainty. The results at the higher density (10^{13} atoms/cm^3) for the cross fluorescent lines λ = 3639 Å and λ = 4058 Å are in agreement within statistical uncertainty; however, the peak-to-peak separation for the λ = 2833-Å resonance line was very much greater (a factor of 5/3). We do not yet know the cause of this effect.

As the density of lead atoms is increased above

10^{14} atoms/cm^3 the Hanle-effect pattern broadens because of the effects of collision broadening as may be seen in Fig.11. Using the results of Byron, McDermott, and Novick[3] and ignoring the effects of wall collisions, we have

$$n\bar{v}\sigma = \frac{1}{\tau_{obs}} - \frac{1}{\tau_c}$$

where n = density of atoms in the cell

$\bar{v}$ = mean velocity of atoms in the cell

σ = the collision cross section

τ_{obs} = observed lifetime

τ_c = the saturation "coherence-narrowed" lifetime value.

The data give a cross section $\sigma = 1.4(2) \times 10^{-12}$ cm^2 in the range of 2×10^{14} atoms/cm^3 to 8×10^{14} atoms/cm^3, where the density has been estimated from the ideal gas law and vapor pressure-vs-temperature curves. The theory of Byron and Foley[4] predicts $\sigma = 0.96 \times 10^{-13}$ cm^2 for resonant collision broadening although in its present form it is not truly applicable to Hanle-effect broadening.

The results of this theory are shown at densities higher than 10^{13} in Fig. 12, which may be compared with the experimental results in Fig. 11.

The presence of fluorescent lines in lead allows a detailed study of collision broadening of the Hanle-effect linewidth without the complications of backscatter techniques.[5] In contrast to cadmium and zinc, the collision broadening in lead should be of the resonant type because of the fairly large f value.[4] Our oven design will be modified to eliminate angular distortion and to permit higher operating temperatures. These investigations should clarify our understanding of the

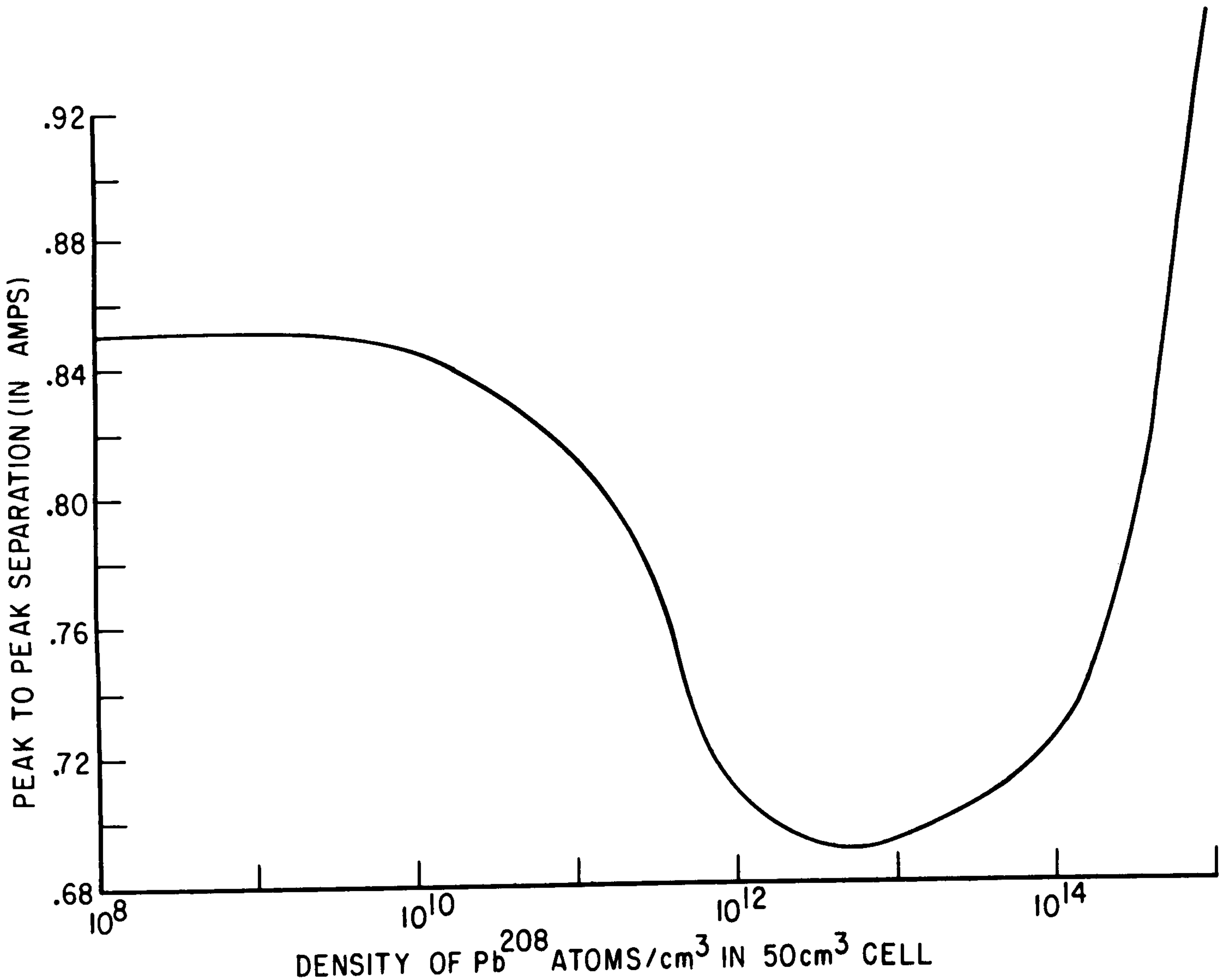

Fig. 12. Theoretical predictions: peak-to-peak separation vs density.

collision mechanisms responsible for disorientations of aligned excited atoms.

The Hanle effect has been observed in the (6p8s) $^3P_1^o$ state by use of the high-field apparatus. By comparing the width observed in the signal from this state with that of the (6p7s) $^3P_1^o$ state, a rough lifetime value $\tau = 7.3(1.4) \times 10^{-9}$ sec has been obtained. More precise measurements in this state will be made in the future.

Preparations are proceeding to determine the hfs of Pb^{205}. Thirty milligrams of 70% separated Pb^{204} will be bombarded in a reactor for three weeks at a neutron flux of 5×10^{14}. Since the thermal neutron cross section of Pb^{204} is 0.8 b, this should result in the production of 20 micrograms of Pb^{205} with a concentration of 10^{-3}. Next, the sample will be isotope separated at an efficiency of about 5% and an enrichment factor of 100. This will leave us with one microgram of Pb^{205} with a concentration of 10% or 3×10^{15} Pb^{205} atoms with 27×10^{15} Pb^{204} atoms. It should be possible to transfer a third of these to an absorption cell giving 10^{15} Pb^{205} atoms. Since our apparatus is capable of detecting as few as 2×10^{10} Pb^{207} atoms in the vapor phase, we should be able to perform hfs measurements on Pb^{205} with this sample.

(1) M. Z. Khokhlov, Contr. Crimean Ap. Obs. 21, 84 (1959); 22, 118 (1960).

(2) J. P. Barrat, J. Phys. Rad. 20, 541 (1959); 20, 633 (1959).

(3) F. W. Byron Jr., M. N. McDermott, and R. Novick, Phys. Rev. 134, A615 (1964).

(4) F. W. Byron Jr., and H. M. Foley, Phys. Rev. 134, A625 (1964).

(5) CRL Quarterly Progress Report, June 15, 1962, p. 15.

III. PHYSICS OF MOLECULES

A. BEAM MASER SPECTROSCOPY*

(P. Cahill, L. P. Gold, P. Thaddeus†)

The hyperfine spectrum of the $3_{03}-3_{13}$ rotational transition in NH_2D has been observed at 43042.48 Mc with a 20 to 1 signal-to-noise ratio for the nitrogen hyperfine lines. A 45V10 OKI klystron was phase locked by use of a Dymec Oscillator Synchronizer to the fifth harmonic of an LFE Ultra-Stable Microwave Oscillator, operated at 8596.6 Mc. The klystron was frequency modulated by sweeping the oscillator synchronizer with the sawtooth from a 502 Tektronix Oscilloscope. Presently the spectrum is being measured by using the forty-third harmonic of the 1000-Mc CRL frequency standard and a Collins receiver. In the next quarter the spectrum will be measured, and the spin-rotation and quadrupole coupling constants of the deuteron will be calculated. This particular transition in NH_2D gives a very precise determination of the deuteron coupling constants since the protons, by virtue of the symmetry of the transition, do not contribute to the multiplicity of the hyperfine structure. Once values of the deuteron coupling constants have been determined, the previously observed hyperfine spectrum of the $3_{03}-3_{13}$ rotational transition at 18807 Mc(1) will be re-analyzed to determine more precisely the proton coupling constants. Also, since it is possible to operate a beam maser at millimeter frequencies, the hyperfine spectrum of the $2_{02}-2_{12}$ rotational transition in NHD_2 will next be investigated at 38739 Mc, and in conjunction with the previously observed spectrum of the $3_{03}-3_{13}$ transition in NHD_2,(2) the hyperfine coupling constants for the deuteron and proton in NHD_2 will be determined.

*This research was also supported in part by the Air Force Office of Scientific Research under Grant AF-AFOSR-330-63, and in part by the Office of Naval Research under Contract Nonr-266(45).

†NASA Institute for Space Studies.

(1) P. Thaddeus, L. C. Krisher, and P. Cahill, J. Chem. Phys. 41, 1542 (1964).

(2) CRL Quarterly Progress Report, June 15, 1964, p. 25.

B. MOLECULAR BEAM VELOCITY SELECTOR*
(M. M. Hessel, P. Kusch)

Previously it had been reported[1] that an inverse fifth-power radial dependence of the interaction potential was observed for the scattering of KCl by N_2 and Ar. The Massey-Mohr theory,[2] which requires that true quantum cross sections be measured, was used to interpret the data. The question has arisen as to whether our apparatus has sufficient angular resolution to produce attenuation characteristic of true quantum cross sections. Massey and Mohr[2] have given a crude criterion for estimating the angular resolution below which true quantum cross sections are obtained. According to this criterion the angular resolution of our apparatus should be better than 450 seconds; it is actually 150 seconds.

Even though the angular resolution of the apparatus is adequate according to this rough criterion, an experimental check should be made to determine the effect of improved resolution.

The resolution of the apparatus was improved to 75 seconds when the size of the entrance slit of the scattering chamber was reduced from 0.008 in. to 0.004 in. KCl-Ar scattering experiments are now in progress.

*This research was also supported in part by the Air Force Office of Scientific Research under Grant AF-AFOSR-330-63, and in part by the Office of Naval Research under Contract Nonr-266(45).

(1) CRL Quarterly Progress Report, December 15, 1964, p. 23.

(2) H. S. W. Massey and C. B. O. Mohr, Proc. Roy. Soc. (London) A144, 188 (1934).

C. MOLECULAR BEAM ELECTRIC RESONANCE SPECTROSCOPY*
(P. Cahill, L. P. Gold)

The object of this program[1] is the measurement of the microwave and rf spectra of simple molecules by the molecular beam electric resonance method.

During this quarter, research in microwave spectroscopy and beam maser spectroscopy limited the experimental program on this project to routine tests.

Program for the next interval: If the rotational spectrum of KCN is observed in the high-temperature microwave spectrometer (see Section III.D of this report), an attempt will be made to observe the same transition in the molecular beam electric resonance spectrometer.

*This research was also supported in part by the Air Force Office of Scientific Research under Grant AF-AFOSR-330-63, and in part by the Office of Naval Research under Contract Nonr-266(45).

(1) CRL Quarterly Progress Report, March 15, 1962, p. 45.

D. MICROWAVE SPECTROSCOPY*
(P. Cahill, L. P. Gold)

Part I

During this quarter, the high-temperature microwave spectrometer has been used to search for the rotational spectrum of potassium cyanide. The chemical behavior of the system has proved satisfactory; potassium and ammonia, produced in an earlier cell of different design[1] by sample decomposition, are not observed in the present system. However, no definite spectral lines have yet been observed.

Program for the next interval: The search for the rotational spectrum of KCN will continue.

Part II

During this quarter, work on the microwave spectrum of methyl vinyl ether has been concentrated mainly on obtaining more information about the *cis* form. Accurate measurements of a number of the previously assigned lines have been made both in this laboratory and by Dr. Noel Owen of Harvard University in collaboration with whom this project is being carried out. Careful observation of these lines failed to reveal an observable splitting. A search was therefore made for the rotational spectrum of excited vibrational states. One such state has been identified; the resulting rotational constants are given in Table I together with the previously obtained ground-state constants. If this excited state is due to a methyl group torsional vibration, it may be possible to obtain a value for the barrier to internal rotation of the methyl group despite the absence of observable internal rotation splitting in the spectral lines.

TABLE I. Rotational constants of *cis* methyl vinyl ether in Mc.

	Ground State	Excited Vib. State
A	18224.96	18301.52
B	6388.96	6331.44
C	4875.85	4861.80
κ	-0.773302	-0.781299

Program for the next interval: Work will continue on the measurement and analysis of excited state spectra.

*This research was also supported in part by the Air Force Office of Scientific Research under Grant AF-AFOSR-330-63, and in part by the Office of Naval Research under Contract Nonr-266(45).

(1) CRL Quarterly Progress Report, June 15, 1963, p. 32.

E. MAGNETIC ROTATION SPECTRA*
(P. Kusch, B. Palatnick)

Work is continuing toward the improvement of the quality of the spectrograms.[1] Some experimentation is being carried out using a 10-in. absorption tube, which is half the length of the previous one. The remaining space inside the magnet is filled with iron plugs, giving a higher average field over the length of the tube.

Program for the next interval: More spectrograms will be taken until the quality of the data is satisfactory. At that point, reduction and analysis of the data will begin.

*This research was also supported in part by the Air Force Office of Scientific Research under Grant AF-AFOSR-330-63, and in part by the Office of Naval Research under Contract Nonr-266(45).

(1) CRL Quarterly Progress Report, December 15, 1964, p. 26.

A. INTERACTION BETWEEN A NEUTRAL BEAM AND A CONDUCTING SURFACE*
(P. Kusch, D. Raskin)

The pressure-dependent scattering background reported previously[1] was still evident in data taken during the last interval. Attempts were made to improve the ultimate pressure of the vacuum system. The lowest pressure measured was 5.4×10^{-9} Torr. Several unsuccessful attempts were made to seal the existing flanges with aluminum or gold gaskets.

At present, the possibility of building a new vacuum system is being explored in order to obtain a pressure of about 1×10^{-10} Torr.

Program for the next interval: Different pumping systems will be studied and evaluated. At the same time experiments will be done on the present system in order to understand completely the observed beam profile so that all necessary improvements can be made in the design of a new system.

*This research was also supported by the Army Research Office (Durham) under Grant DA-ARO(D)-31-124-G568.

(1) CRL Quarterly Progress Report, December 15, 1964, p. 34.

B. ENDOR AND OPTICAL STUDY OF COLOR CENTERS*
(I. Bass, M. Dakss, D. Daly, A. N. Jette, R. Marzke, R. Mieher, S. C. Ribeiro)

ENDOR measurements have been completed for the H-center in lithium fluoride, and the data are now being analyzed. A detailed discussion on the experiment and its results will appear in the next quarterly report.

The dipolar contribution to the magnetic hyperfine constants at the surrounding nuclei for the V_K-center in LiF was calculated using the Hartree-Fock wave functions of A. C. Wahl for F_2^-.[(1)] Values were obtained for several internuclear distances for the molecule-ion and relaxed positions of the surrounding nuclei. Good agreement was found between the experimental constants determined by ENDOR measurements in this laboratory and the calculated values using an internuclear distance of 3.8 atomic units for the molecule-ion. An attempt will be made to calculate the contact interaction at the surrounding ions to the V_K-center including the influence of exchange polarization. Lattice energy calculations are in progress to assess the relaxations of neighboring ions. A more detailed account will appear in a subsequent quarterly report.

*This research was also supported by the National Science Foundation under Grant NSF-GP 3385.

(1) A. C. Wahl, unpublished work at the Argonne National laboratory.

C. ADIABATIC DEMAGNETIZATION IN THE ROTATING FRAME*
(H. Einbinder, S. R. Hartmann)

Work continued on the construction of the He^3 cryostat. Requirements were determined for the He^3 circulating pumps, and filters were designed for the receiver used to detect the signal from the He^3 nuclei.

Further experimentation was performed to determine the effect of irradiation of the F^{19} nuclear spin system in CaF_2 by a radio-frequency field near resonance. The time required for the spin system to reach thermal equilibrium with the crystal lattice (spin-lattice relaxation time) was measured for both an initial state consisting of a completely disordered system and an initial state produced by an adiabatic demagnetization in the rotating frame (ADRF). The decision was made to obtain a CaF_2 crystal with an impurity content producing a spin-lattice relaxation time more convenient for the experiment.

<u>Program for the next interval</u>: Investigation will continue of the characteristics of the state produced by ADRF performed on the F^{19} nuclear spin system. Completion of the He^3 cryostat is anticipated.

*This research was also supported by the National Science Foundation under Grant NSF-GP 3379.

D. HIGH-FREQUENCY PROPERTIES OF SUPERCONDUCTORS*
(S. Zemon)

Calculations have begun for the determination of the predictions of the Bardeen-Cooper-Schrieffer theory(1) for the surface resistance of zinc.

<u>Program for the next interval</u>: The calculations will be completed and compared with our surface-resistance data for zinc.

*This research was also supported in part by the National Science Foundation under Grant NSF-GP 1031 and in part by the Office of Naval Research under Contract Nonr-3994(00).

(1) J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. <u>108</u>, 1175 (1957).

E. ELECTRONIC TUNNELING IN THE SUPERCONDUCTING STATE*
(S. Zemon)

In order to generate higher electric fields in our thin-film junctions, considerable effort went into producing films thinner than 1000 Å. Films with thicknesses of 100 Å or less either were not electrically continuous or they disintegrated rapidly in air. This was true for films deposited upon a substrate which was at a temperature of -100°C. Furthermore, over these films was a protective coating of SiO. Film widths were 0.010 in. and initial junction resistances were between 1 and 100Ω. The deposition of both films was executed without disturbing the evaporator arrangements. A good aluminum-aluminum junction was fabricated with a total thickness of 600 Å. Microwave radiation in a cylindrical cavity produced a sharp junction heating effect even with 15 dB of microwave attenuation, but no effect similar to the photon-assisted tunneling, reported by Dayem and Martin,[1] was seen at 8 different frequencies between 50 and 60 kMc/sec at temperatures as low as 0.3°K.

It is of interest to compare the energy gap obtained in our surface resistance measurements with that derived from tunneling measurements. Therefore, during this quarter techniques were developed for the deposition of zinc thin films.

Program for the next interval: Zinc and cadmium tunneling characteristics will be measured.

*This research was also supported in part by the National Science Foundation under Grant NSF-GP 1031 and in part by the Office of Naval Research under Contract Nonr-3994(00).

(1) A. H. Dayem and R. J. Martin, Phys. Rev. Letters 8, 246 (1962).

F. NUCLEAR MAGNETIC RESONANCE IN PLATINUM*
(S. R. Hartmann, G. R. Mather)

Platinum samples, consisting of 0-10μ particles, have been prepared and the platinum resonance observed. The platinum particles were suspended in vaseline in order to insulate them electrically, and the mixture was placed in lucite containers, made by boring holes in lucite rods of various diameters.

The resonance was first observed in a field of 3kG at 77°K, and later in the same field but at 4.2°K. Signal-to-noise ratio at the latter temperature is quite good, and little trouble from this source is anticipated in taking data. However, an unstable oscillator in conjunction with the phase-sensitive detection made the signal quite jittery, especially at larger pulse separations. The Hewlett-Packard rf oscillator was therefore replaced with a 5-Mc crystal oscillator which has a short-term stability somewhat better than 1 part in 10^6 and a long-term stability of about 1 part in 10^5. At the same time new pole caps were put on the magnet so that fields up to 6 kG are now possible. These pole caps, however, produce fields a good deal more inhomogeneous than the previous caps, so that free induction decay times of only 50 μsec are observed. The bandwidth of the receiver must, therefore,be larger than for more homogeneous fields, and consequently, the signal-to-noise ratio is not as large as it might be. This situation is currently being corrected; first by the use of a small sample volume, and second by the use of a boxcar integrator with recorder readout.

Calculations of the relaxation times to be expected in the very small particles (50-Å linear dimension) are currently under way. Since platinum is a transition metal with a rather large state density at the Fermi surface, some question arises as to whether it is in fact suited to a study of fine particle effects.(1) In judging whether or not these effects should be expected in a given metal, it is necessary to consider the widths of the electron energy levels relative to their separations. Order-of-magnitude calculations for platinum, based on inferred values for the density of states[(2)] and on the electrical conductivity, indicate that 50-Å platinum particles are at best marginal for such studies. The decision has therefore been made to look for small particle effects in the platinum sample experimentally; if none should arise, then other colloidal metals will be sought.

Program for the next interval: Measurements of the relaxation time T_1 as a function of temperature will be made on the bulk (0-10μ) particles from 4.2°K to lower temperatures. Also, a sample consisting of the 50-Å particles will be prepared, and relaxation time measurements will be taken in this same temperature range.

*This research was also supported by the National Science Foundation under Grant NSF-GP 3379.

(1) CRL Quarterly Progress Report, December 15, 1964, p. 39.

(2) M. Shimizu and A. Katsuki, J. Phys. Soc. Japan 19, 1135 (1964).

V. OPTICAL AND MICROWAVE MASERS

A. INFRARED AND OPTICAL MASERS

1. Optical Maser Spectroscopy

a. Light-Scattering Homodyne Spectroscopy*
(S. S. Alpert, D. A. Balzarini, E. Lipworth, L. Seigel, Y. Yeh)

Work accomplished during this quarter was primarily concerned with obtaining experimental data relating to critical opalescence of a binary system of cyclohexane-aniline near the critical temperature. Further developmental work has been carried out resulting in the construction of a variable volume pressure cell to be used in critical opalescent observations of CO_2 and xenon. An argon laser designed to be run for long periods of time has also been constructed to be used as a light source in future experiments. A novel feature of the argon laser is the manner in which the tube is air cooled. A special cooling arrangement was required to avoid microphonics which could introduce detrimental spurious phase modulation into the operation of the laser homodyne spectrometer. The topics of this quarter are discussed below.

[1] <u>Observation of Time-Dependent Concentration Fluctuations</u>. Using a He-Ne laser homodyne spectrometer,(1) we have observed for the first time the frequency spectrum of the time-dependent concentration fluctuations in a binary liquid mixture just above the critical temperature.

The system studied was a cyclohexane-aniline mixture, 53% cyclohexane and 47% aniline by weight. The mixture was contained in a cylindrical glass cell in a constant-temperature enclosure, the temperature of which could be adjusted and held constant to about 1×10^{-3}°C over extended periods of time.

The frequency spectrum was observed for various temperatures above the critical temperature. Some representative spectra are shown in Fig. 13. As the sample approached its critical temperature T_c from above, a definite narrowing of the width of the scattered light was observed as shown in Fig. 14 where the scattering angle was 20.5° from the forward direction. The linewidth at a particular temperature and scattering angle was found to be independent both of the sample cell thickness (3, 6, and 10 mm) and of the intensity of the laser beam, indicating that multiple scattering and heating effects play only a minor role. The lines were quite well fitted by a Lorentzian curve as anticipated.

We have also investigated the dependence of linewidth on scattering angle. Van Hove(2) has constructed the space-time correlation function for a pure liquid near its critical point and predicted that the shape of the scattered line due to density fluctuations is Lorentzian with a half-width proportional to K^2 where $|\vec{K}|=|\vec{k}_f-\vec{k}_o|$ and where $\vec{k}_o$ and $\vec{k}_f$ are the propagation vectors of the incident and scattered waves respectively. Consequently, the width should be proportional to $\sin^2\theta/2$, where θ is the forward scattering angle. We have verified that this relation holds for concentration fluctuations, and the results are shown in Fig. 15.

[2] <u>Variable Volume Pressure Cell</u>. A variable volume pressure cell has been designed and constructed. The main feature of this assembly is the incorporation into the design of a compressible stainless-steel bellows. This feature allows the volume of the cell to be varied over a dynamic range of about 25%.

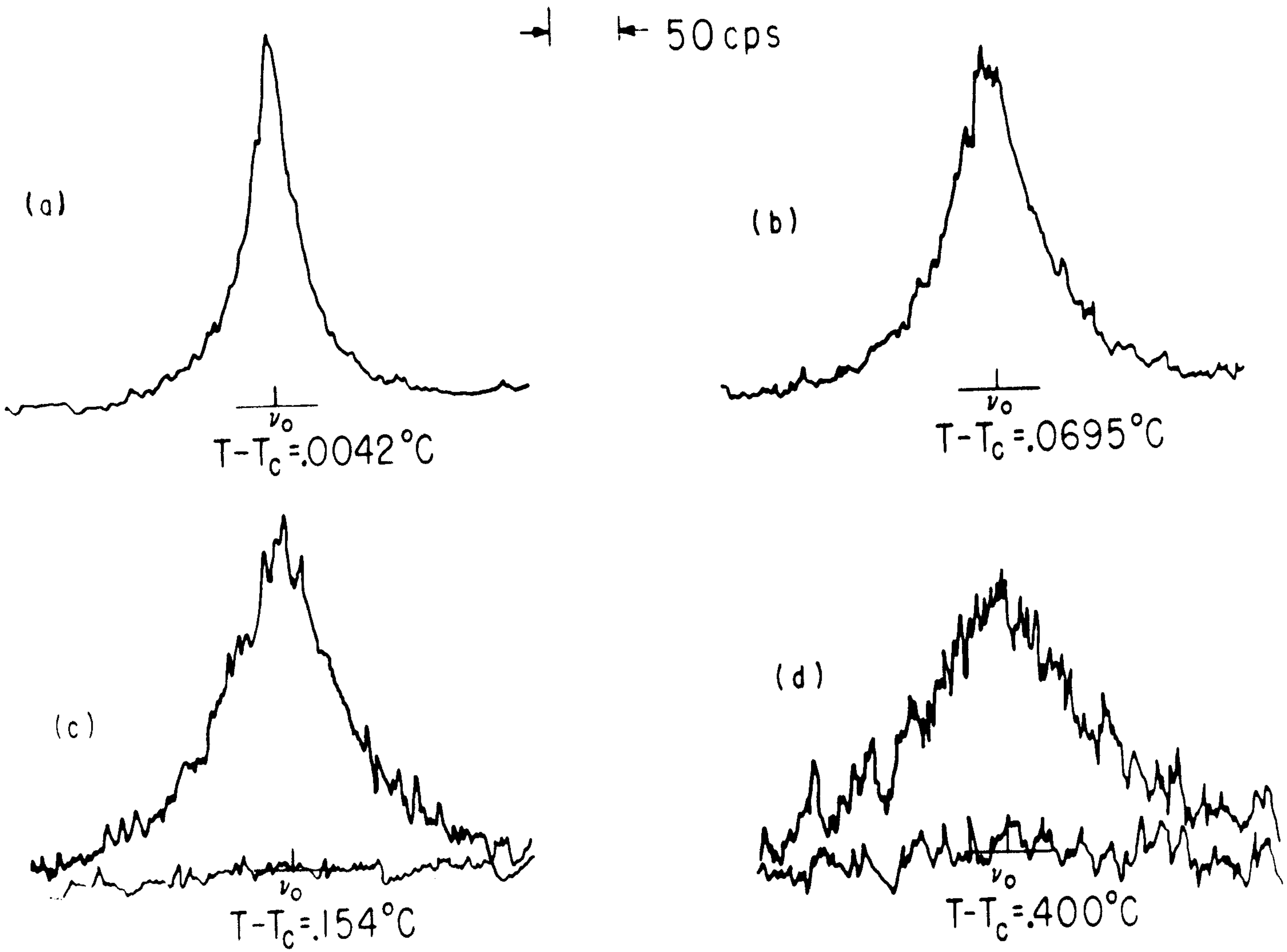

Fig. 13. Spectral profiles.

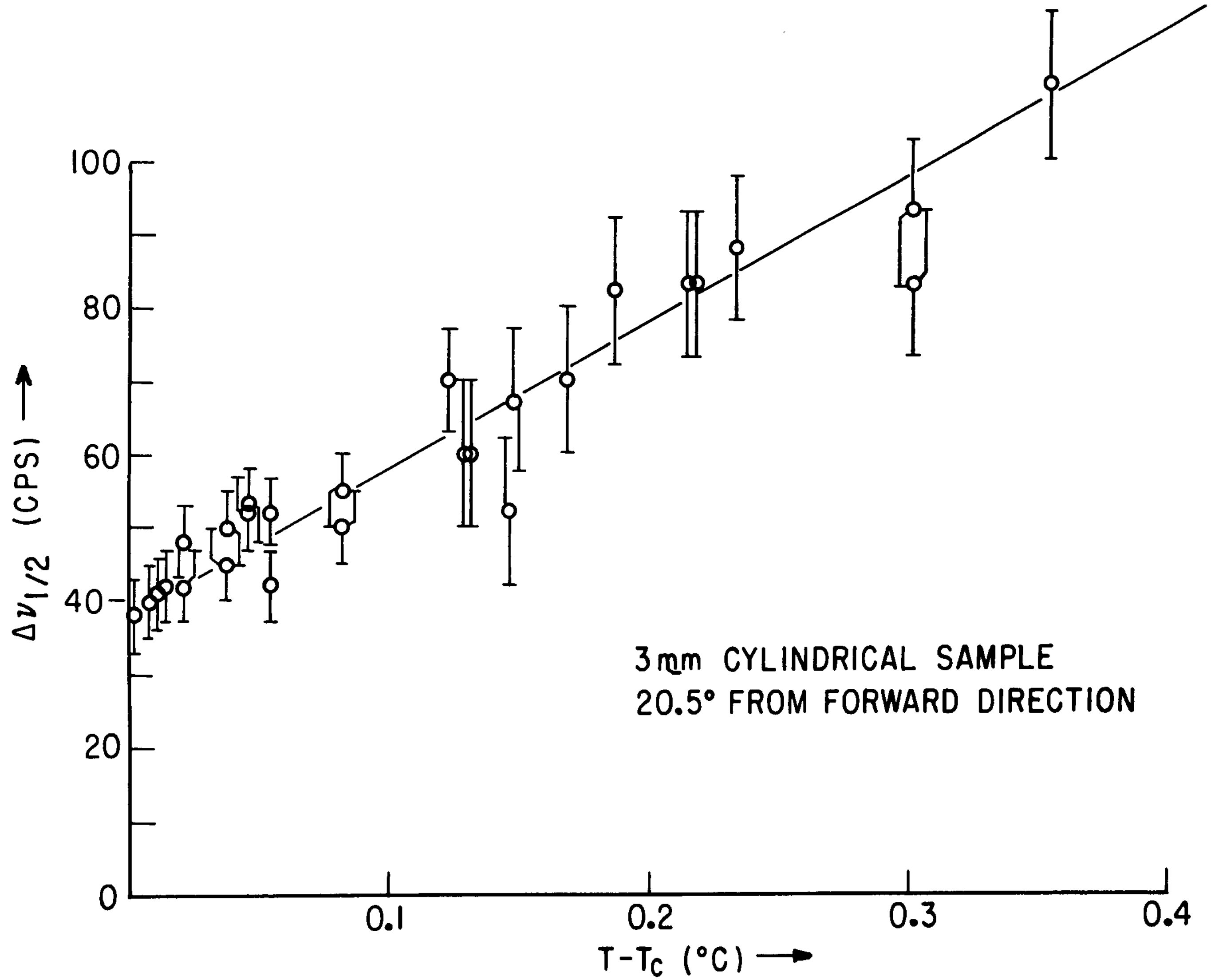

Fig. 14. Width at half intensity vs $T-T_c$. Note that at $T=T_c$, linewidth does not vanish.

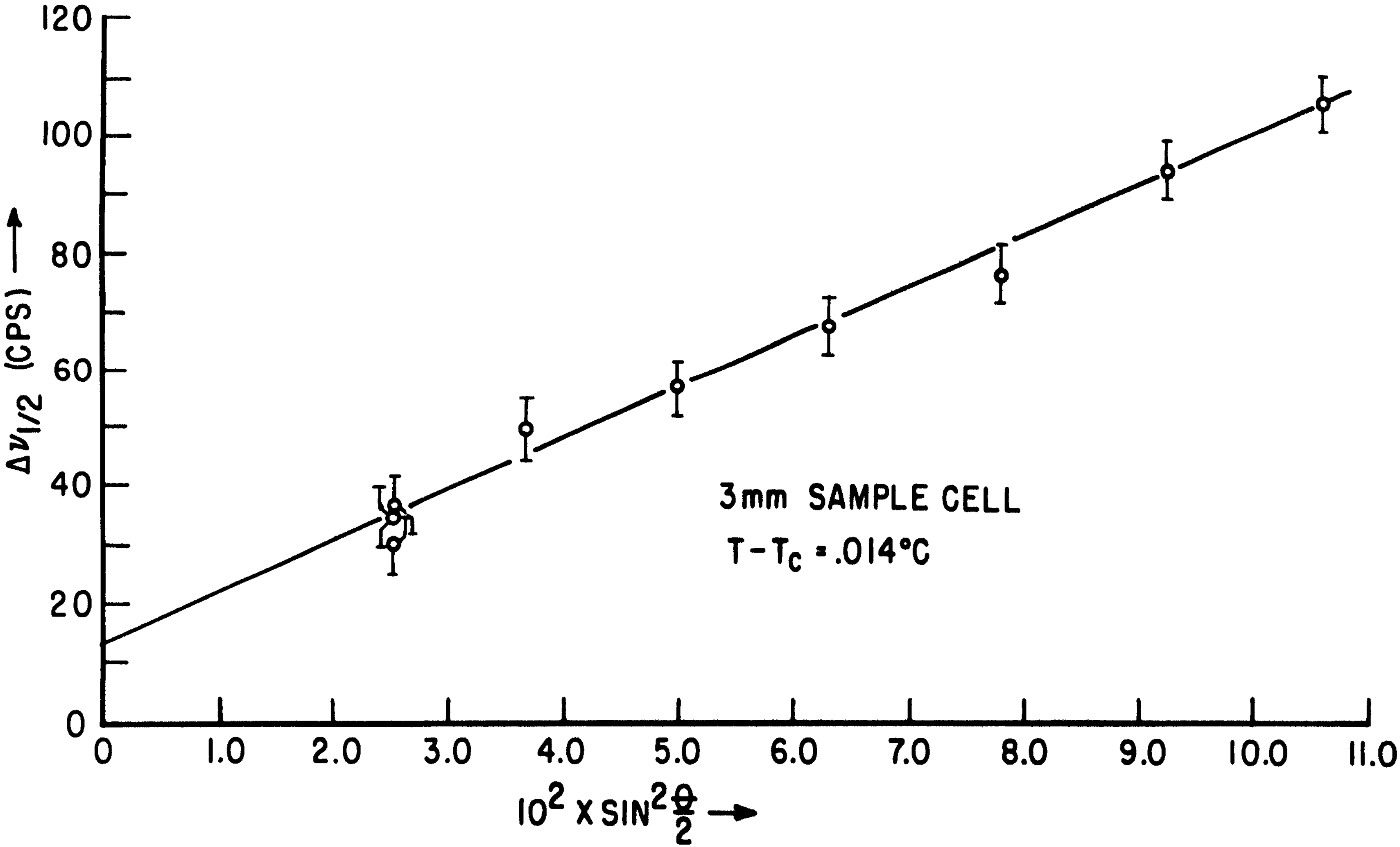

Fig. 15. Width at half intensity vs $\sin^2\theta/2$.

In order to reach the critical point of CO_2 or xenon in a fixed volume cell, it becomes necessary to load such a cell with exact quantities of the experimental substance at temperatures removed from the critical temperature. This is a difficult task; however, the variable volume pressure cell makes it possible to adjust the over-all density of the material so that the critical density can be achieved.

[3] Argon Laser. A workable, stable, and hopefully, long-lived argon ion laser has been constructed. This laser has not as yet been tested to its full power capability but has been operated at outputs of about 30 mW at 4880 Å for long periods of time. The argon laser has a coaxial air-flow shield (Fig. 16) which allows for filament cooling without the use of motor driven fans. Such fans must be avoided because of the microphonics that they introduce.

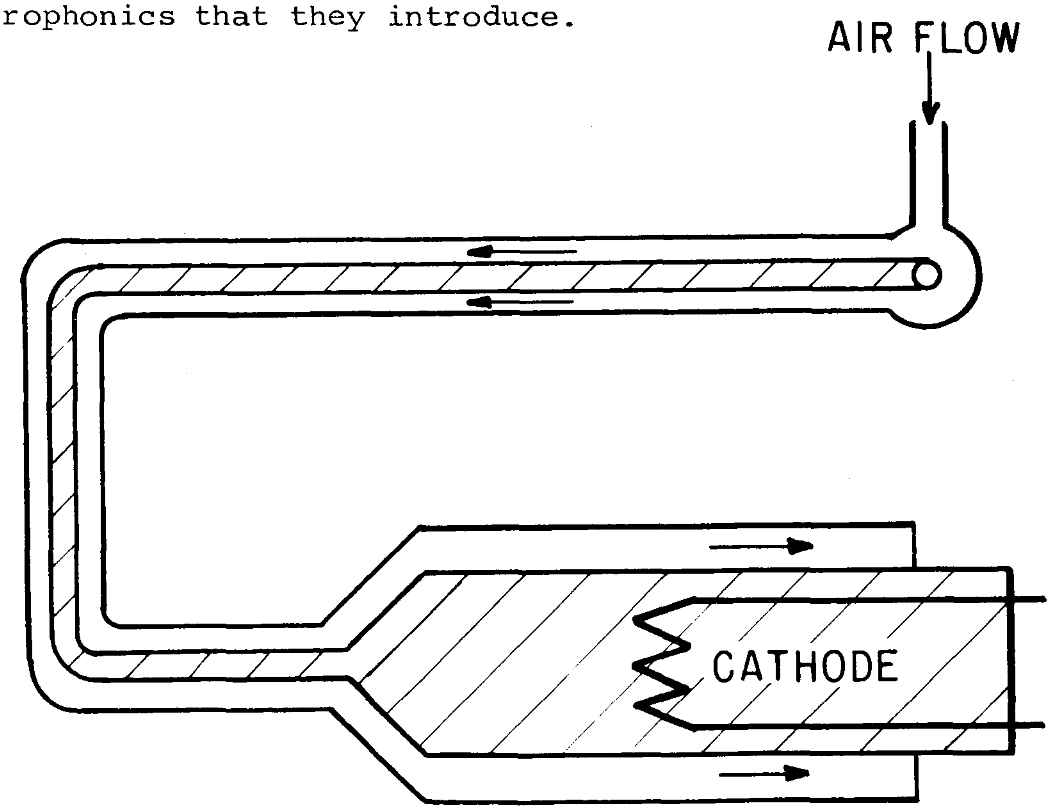

Fig. 16. Schematic diagram of air-cooling shield.

Program for the next interval: Attempts will be made during the next quarter to observe critical opalescence in carbon dioxide and in xenon. The feasibility of the argon laser will be tested and if successful, this tool will possibly be used to extend the temperature range of observation of the cyclohexane-aniline system.

*This research was also supported by the Army Research Office under Contract DA-31-124-ARO-D-296.

(1) H. Z. Cummins, N. Knable, and Y. Yeh, Phys. Rev. Letters 12, 150 (1964)
(2) L. Van Hove, Phys. Rev. 95, 249 (1964)

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b. Laser Studies of Molecular Birefringence*
(R. L. Disch, S. Golub)

[1] Birefringence Induced by an Inhomogeneous Electric Field. Preliminary measurements have been made on gaseous samples of N_2 to check the stability of the apparatus, and measurements on CCl_4 are now in progress.

[2] A Laser Polarimeter. The construction of this device is continuing. A further discussion of it will be given in the next report.

[3] Magnetic Birefringence in Fluids. The construction of equipment for the measurement of the Cotton-Mouton effect in liquids and possibly certain gases has been started. Of special interest is the case of paramagnetic substances, especially transition metal complexes, but attention will also be given to the use of this method in the elucidation of structural parameters in large molecules which can exist in a helical form.

*This research was also supported by the Army Research Office under Contract DA-31-124-ARO-D-305.

2. Ruby Laser: Photon-Echo Resonance*
(I. D. Abella, S. R. Hartmann, N. A. Kurnit)

The effect of magnetic field upon the photon echo has been further studied. As discussed in the preceding Quarterly Progress Report, a magnetic field directed along the optic axis is needed in order to observe photon echoes. It has been found that the necessary magnitude and sensitivity to alignment of this field are dependent upon the time separation of the excitation pulses. As this separation is decreased, it becomes possible to observe echoes at smaller values of field and at larger angles between the field and the optic axis. At 36-nsec pulse separation, echoes begin to be observed at ≈ 7 G and rapidly increase in amplitude with field up to ≈ 50 G, after which no further increase is seen. At this pulse separation, echoes have been observed with the magnetic field at angles of up to 70° from the optic axis.

In Fig. 17 is shown the variation of echo intensity with angle θ between the magnetic field and the optic axis, for two different values of pulse separation τ'. The solid lines are Gaussian curves drawn through the half-widths of the experimental points. For large pulse separations (and consequently for small angles) a fairly good fit to a Gaussian is obtained, whereas a non-Gaussian tail is obtained at shorter times. A plot of the half-width as a function of $1/(\text{pulse separation})^2$ yields a linear relationship (Fig. 18). For small angles the echo intensity is therefore described by the relationship $I = I_o \exp[-k\theta^2\tau^4]$.

The following theory has been proposed to account for the type of relaxation described. The aluminum nuclei in the surrounding Al_2O_3 lattice have a spin of 5/2 which interacts rather strongly with the spin of the chromium ion via an

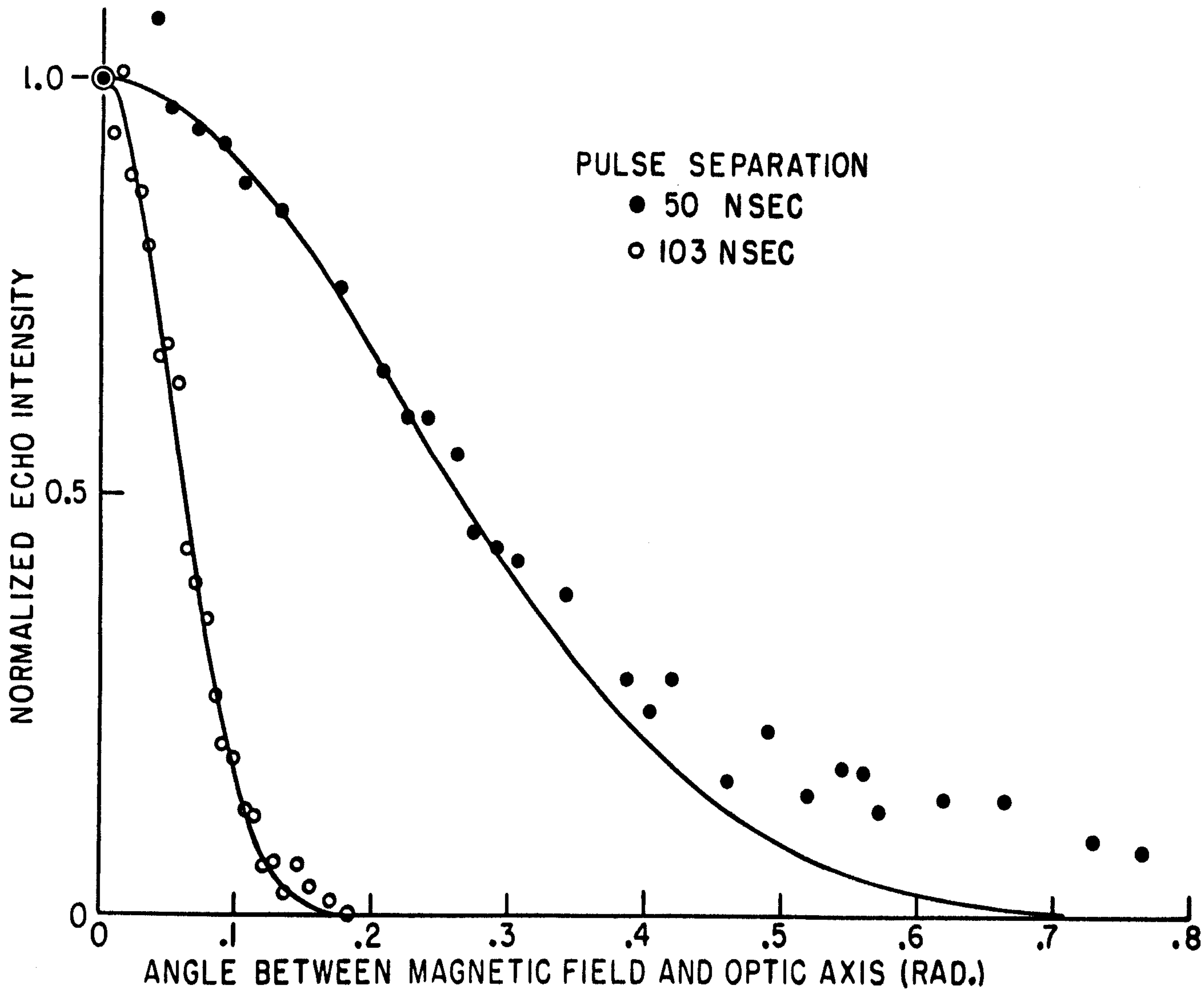

Fig. 17. Echo intensity as function of magnetic field angle.

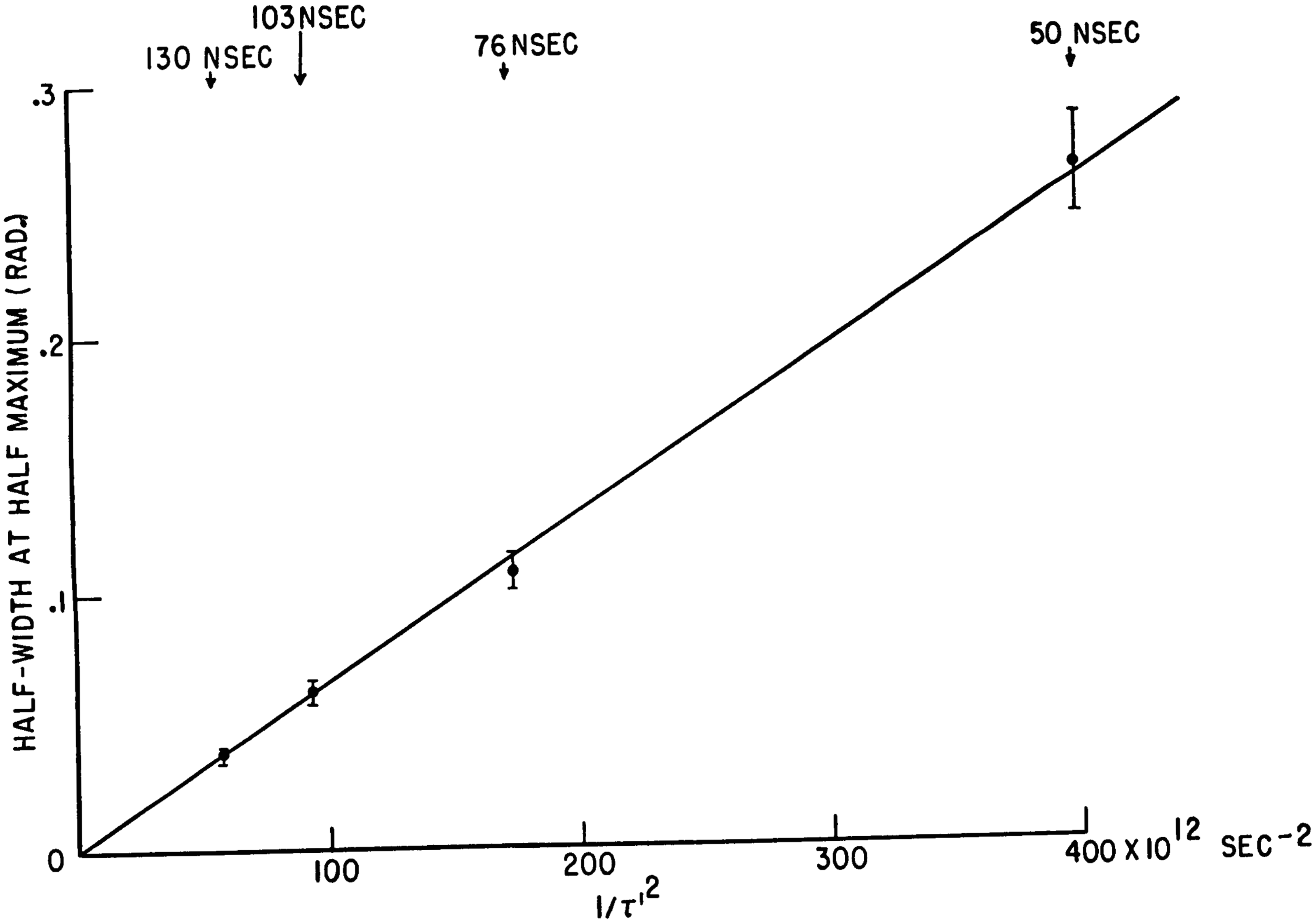

Fig. 18. Angular half-width as function of pulse separation τ'.

exchange interaction transmitted by the oxygen-bonding electrons. This exchange interaction is responsible for an observed 12-G linewidth in the chromium paramagnetic resonance spectrum and also for an effective field of several kilogauss at the aluminum sites.[(1)] In an external field of a few hundred gauss, which makes some angle θ with the optic axis, the chromium electronic spin will be aligned along the direction of the magnetic field since the $^4A_2$ ground state has a nearly isotropic g factor. The aluminum nuclei surrounding chromium sites will be polarized in the effective field of the chromium ion since this is much larger than the applied field. If the chromium ion is now placed in a superposition of ground and excited states by the application of an optical pulse, the direction of its magnetic moment will be abruptly changed because the g factor of the excited state is highly anisotropic and allows the spin to be aligned only along the optic axis. The effective fields at the various aluminum nuclei are therefore abruptly changed in direction, and the aluminum spins begin to precess about these new directions. These precessing spins in turn produce a modulation of the energy levels of the chromium ion; that is, the energy difference between the excited and ground states becomes a function of time: $\Delta E = \Delta E(t)$. The formation of a photon echo is dependent upon the phase increment gained in the interval $(0,\tau')$ being cancelled by that lost in the interval $(\tau',2\tau')$, and this condition is no longer exactly met:

$$\Delta\varphi = \int_0^{\tau'} \frac{\Delta E(t)}{\hbar}\,dt - \int_{\tau'}^{2\tau'} \frac{\Delta E(t)}{\hbar}\,dt \neq 0 .$$

Although it is difficult to treat this problem exactly because of the large number of neighboring aluminum nuclei, the adoption of several simplifying assumptions leads to a relaxation of the form and order of magnitude experimentally observed.

Several crystals of varying concentration have been used to obtain photon echoes in addition to the original 0.005% crystal. Echo intensity appears to increase approximately linearly between 0.005% and 0.05% indicating that there may be some radiation damping effects. Exact comparison of the relative intensities is difficult because the echo-intensity may depend on other factors as well, such as local variations in the direction of the optic axis. Echoes have been obtained in a 0.5% crystal, with about the same intensity as in the 0.05% crystal. No concentration effects were observed on the magnetic-field dependence.

In addition, photon echoes have been obtained with the two excitation pulses traveling in the same direction. A second Kerr cell was needed behind the sample to attenuate the excitation pulses sufficiently. A by-product of this experiment was the observation of two "secondary" echoes caused by the interaction of the echo with the preceding pulses.

A new conduction-cooled tail section has been constructed for the helium sample dewar to allow measurements above $4.2^{\circ}K$. This will be used to study relaxation times due to phonon interactions with the excited state.

<u>Program for the next interval</u>: A measurement will be made of the phonon-induced relaxation time, and further magnetic-field and concentration-effect studies will be carried out.

*This research was also supported by the Army Research Office under Contract DA 31-124-ARO-D-224.

(1) N. Laurence, E. C. McIrvine and J. Lambe, J. Phys. Chem. Solids 23, 515 (1962).

B. RUBIDIUM MASER*

(P. Davidovits and W. A. Stern)

During this quarter the effect of optical pumping on the frequency of an optically pumped Rb^{87} maser oscillator has been investigated. The pumping light causes a shift in the ground-state hyperfine energy levels of Rb^{87} and a corresponding change in the oscillator output frequency. The magnitude and sign of the frequency shift are a function of the pumping light intensity and of the spectral profile of the pumping light.[1]

If the light is centered on the long-wavelength side of the atomic transition, the atomic level will be shifted to a lower energy (case A). If the light is centered on the short-wavelength side of the transition, the level will be shifted to higher energy (case B). This is shown schematically in Fig. 19.

In case A [light centered on the long-wavelength side of the $^2 5S_{\frac{1}{2}}$(F=1) → 5P transition] the F=1 level will be shifted downward, and the maser transition frequency will increase. In case B [light centered on the short-wave length side of the $^2 5S_{\frac{1}{2}}$(F=1) → 5P transition], the F=1 level will be shifted upward, and the maser frequency will decrease. In both cases, it is assumed that there is negligible intensity at the $5S_{\frac{1}{2}}$ (F=2) → 5P transition frequency so that the F=2 level is not pumped.

The frequency displacement of the pumping light with respect to the Rb^{87} absorption lines has been determined by spectroscopic measurements, and the corresponding change in the oscillator frequency was measured. The measurements were made as a function of the light source parameters. The sign and magnitude of the change in the oscillator frequency are consistent with the observed light profiles. The displacement of the pumping light with respect to the Rb^{87} absorption lines is

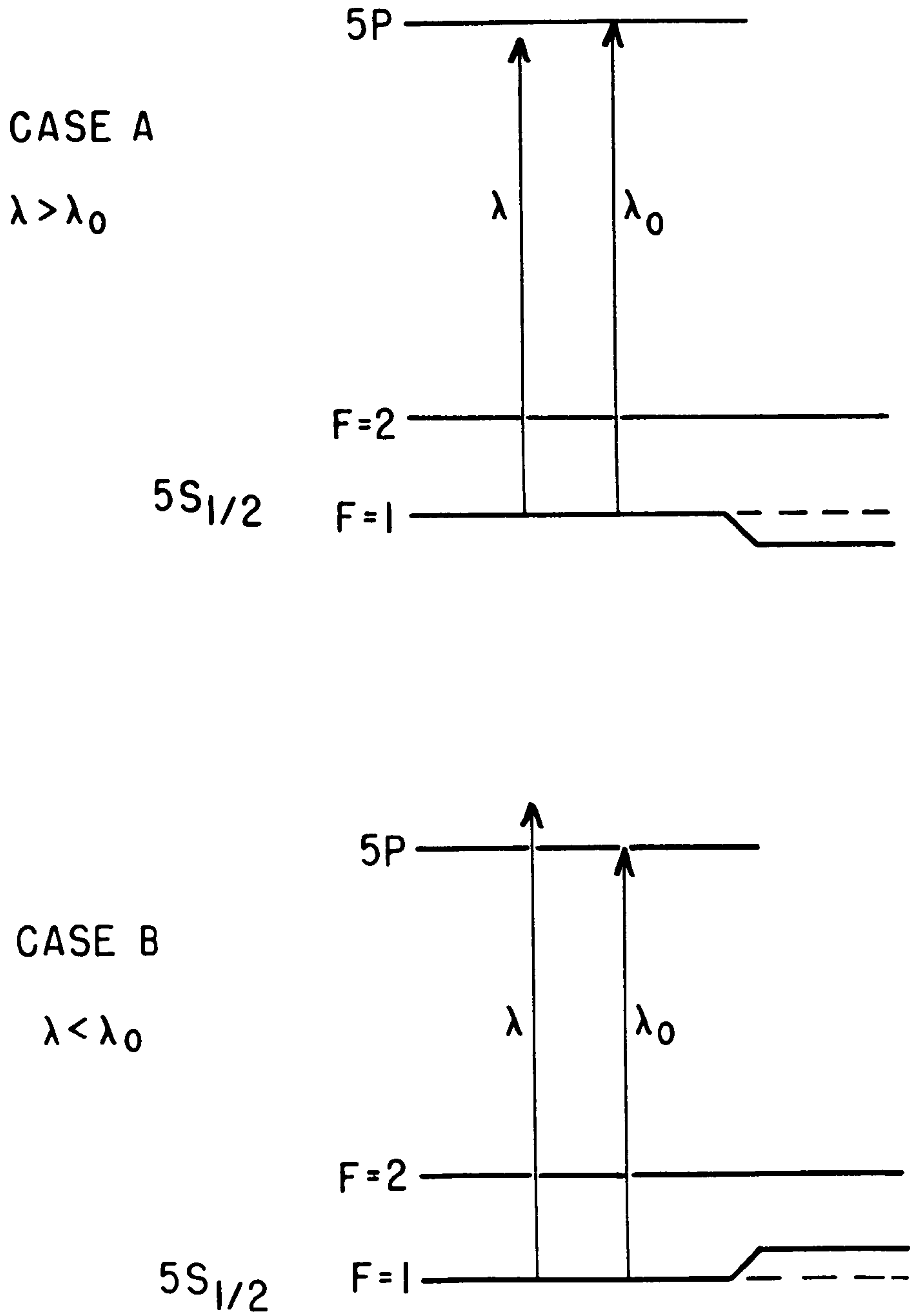

Fig. 19. Hyperfine transition frequency shifts due to optical pumping.

caused by the nonsymmetric absorption of the (5S, F=1 → 5P) Rb^{87} pumping light resulting from the partial overlap with the (5S, F=2) Rb^{85} absorption line in the Rb^{85} filter cell. (The Rb^{85} filter cell is an integral part of the pumping source.) The interaction displaces the pumping light toward the shorter wavelength thereby causing a consistent decrease in the maser oscillation frequency.

Since the pumping light dependence of the oscillator frequency is a source of instability, it is desirable to eliminate the dependence. It is suggested that the 5S, F=2 level may be depopulated and the nonsymmetric absorption thus eliminated by optically pumping the Rb^{85} filter cell. A suitable source for pumping the Rb^{85} filter consists of a Rb^{85} discharge lamp followed by a Rb^{87} filter cell. By this technique the displacement of the maser pumping light and the resulting change in the oscillation frequency may be eliminated.

Also experiments indicate that it is possible to "tailor" the lamp profile by using a natural rubidium discharge pumping lamp. At present measurements are being made to correlate quantitatively the lamp profiles with the maser output frequency. These results will be presented in the next report.

The equipment for studying the quenching properties of buffer gases is nearly completed.

Program for the next interval: 1) The investigation of the optical pumping process will continue. 2) Studies of the quenching properties of buffer gases will begin. 3) The redesigned vacuum-tight cavity will be assembled for further experiments.

*This research was also supported by the Office of Naval Research under Contract Nonr-4259(10).

(1)A. Kastler, J. Opt. Soc. Am. 53, 902 (1963).

A. MODELS OF PLANETARY ATMOSPHERES*

(W. Ho, I. Kaufman, P. Thaddeus†)

Preliminary analysis of the microwave absorption data obtained by the tunable cavity method has indicated a need for extending the temperature range in which measurements are made and for improving the sensitivity of the experimental apparatus. Absorption measurements were made at 9.26 Gc/sec on gas mixtures containing from 5% to 100% carbon dioxide in nitrogen and in argon. The measurements were repeated at approximately 30°C intervals from -40°C to +160°C. Measured absorption coefficients at the high temperatures approached the limit of sensitivity for the low carbon dioxide concentration mixtures. A reliable determination of the temperature dependence of absorption, however, requires a wider range of temperatures than has been used so far.

The extension of the operating temperature to what should be at least 300°C was achieved by replacing the asbestos packing around the tuning plunger shaft with a flexible bellows assembly, which permitted the plunger to be completely sealed within the cavity. The bellows were about ¼ in. in diameter and are made of monel metal. They have so far been tested up to 1800 psi without any leaks. A high-temperature silicone fluid is used in a thermistor-controlled heat bath, and the temperature is monitored with thermocouple junctions attached to the cavity.

To increase the sensitivity of the apparatus work has been concentrated on improving the accuracy of reading the commercial variable attenuator which is used both to measure coupling and to set the half-power level. Attenuation is achieved in this rotary vane device by rotation of a section of cylindrical waveguide which contains a sheet of absorbing material, and is a function of the angle through which the section has turned. The present arrangement now contains an optical system whereby the angle is measured by the deflection of a beam of light, rather than by the mechanical gear system provided with the attenuator.

Program for the next interval: The remaining measurements at temperatures up to 300°C are expected to be completed during the next report period.

*This research was also supported by the National Aeronautics and Space Administration under Grant NsG-442

†NASA Institute for Space Studies

A. POLARIZATION MEASUREMENTS
(R. C. Isler, A. Lurio, R. Novick)

A dual section copper container for forming a solid hydrogen scattering target has been completed (Fig. 20). The lower chamber will hold liquid helium for the refrigerant; the upper perforated section will be lined with ½ mil mylar and used as the hydrogen container. Techniques for bonding the mylar to the copper walls are being explored at the present time.

A second approach is being considered for the target construction. Liquid hydrogen under approximately three atmospheres pressure would be used. The gas which boiled off would be expanded through a nozzle to maintain a temperature of about 22°K. A design of this type has the advantage of not requiring liquid helium.

An annular geiger counter has been designed to fit around the scattering cylinder and is now under construction. This counter has sixteen separate discharge chambers to measure the angular distribution of the scattered x rays.

<u>Program for the next interval</u>: The feasibility of making both solid and liquid hydrogen targets will be explored.

Fig. 20. Hydrogen container in the abstract.

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